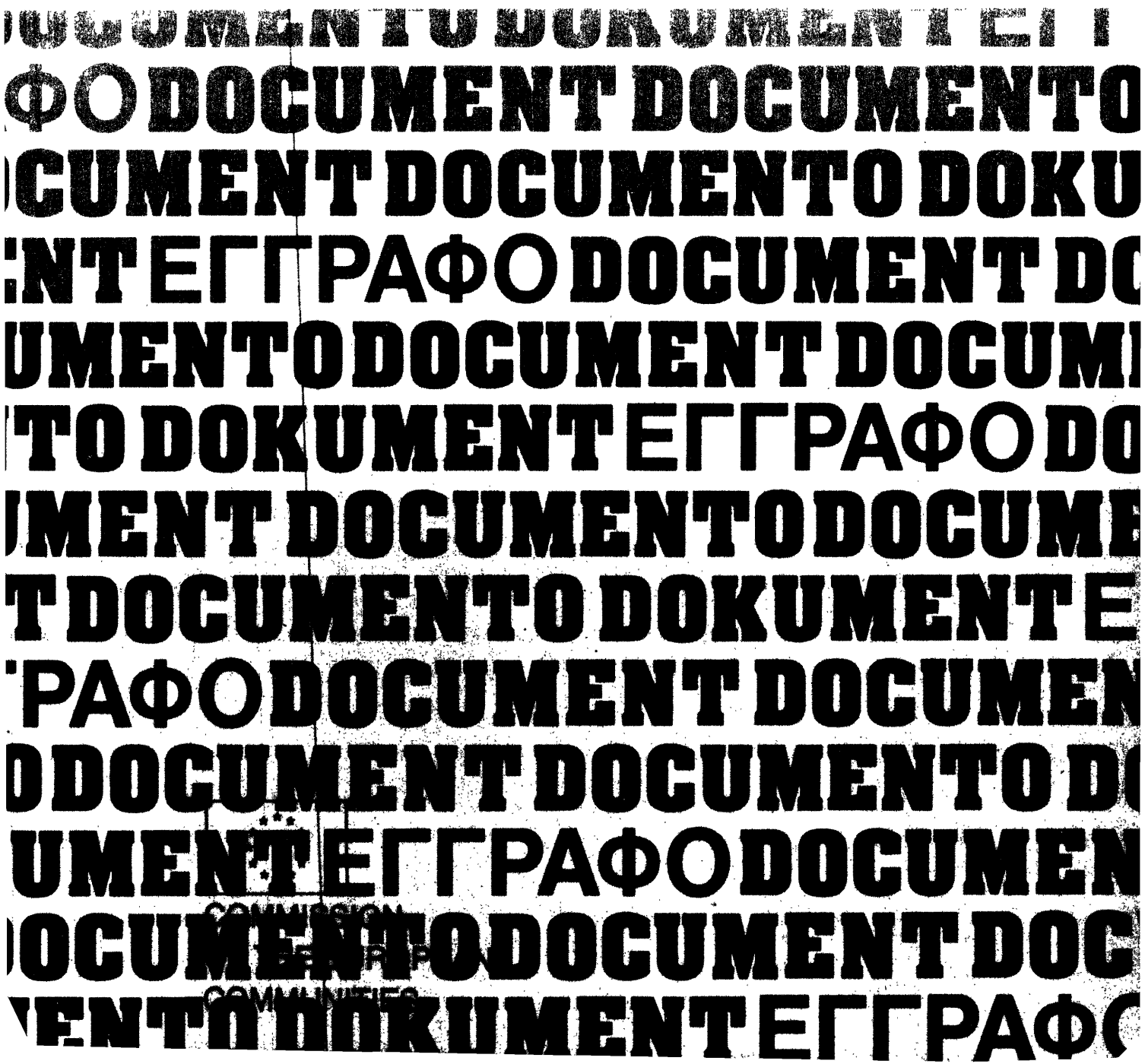
UPDATING OF DATA CONCERNING THE IMPACT ON THE AQUATIC ENVIRONMENT OF CERTAIN DANGEROUS SUBSTANCES, SECOND PART

Part I — Benzene, ethylbenzene and isopropylbenzene



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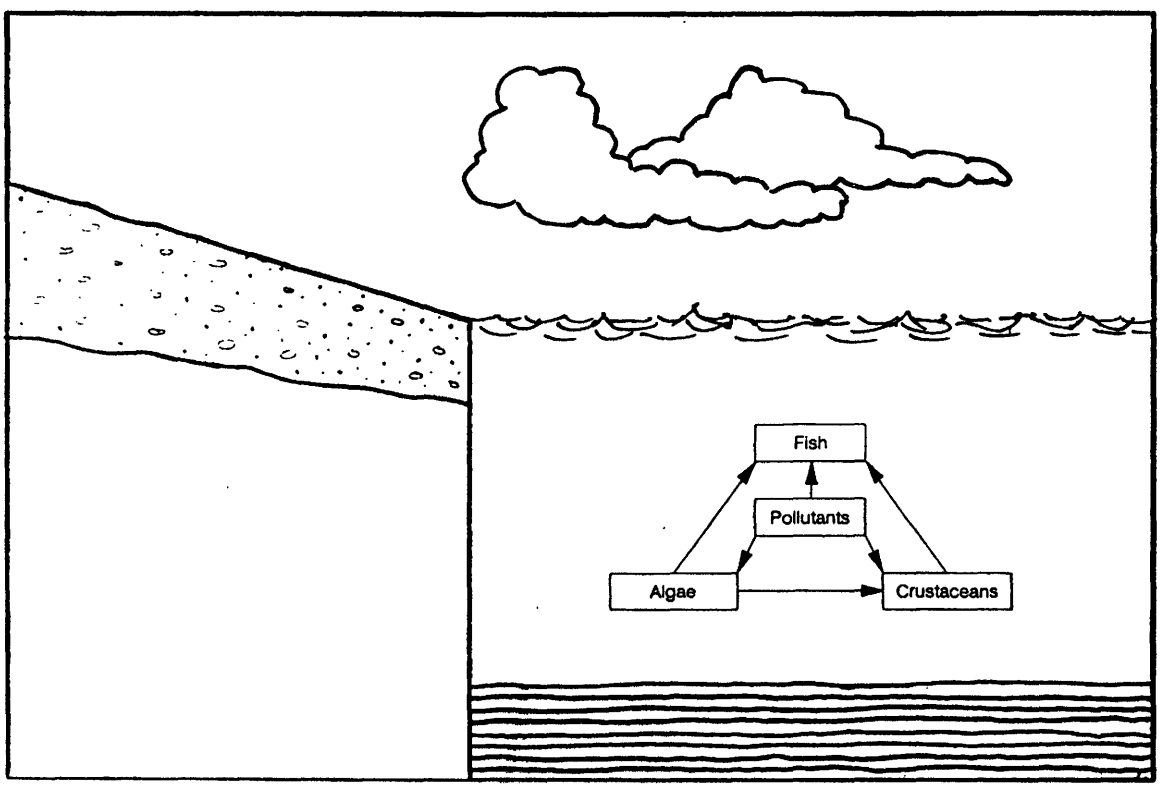
XI/A/3

Updating of data concerning the impact on the aquatic environment of certain dangerous substances, 2nd part

Part I Benzene, ethylbenzene and isopropylbenzene



May, 1992

bkh consulting engineers

PREFACE

The Commission of the European Communities, Directorate-General for Environment, Nuclear Safety and Civil Protection has commissioned BKH Consulting Engineers to carry out Study Contract B6612-90-006688:

> Updating of data concerning the impact on the aquatic environment of certain dangerous substances, 2nd part: benzenes.

The study deals with the following dangerous substances which belong or should belong to List I of Directive 76/464/EEC:

- benzene (7)
- ethylbenzene (79)
- isopropylbenzene (87)
- chlorobenzene (20, 53, 54, 55, 109, 117, 118)
- chloronitrobenzene (27, 28, 29, 30, 63)
- chloroanilines (17, 18, 19, 52)

The study updates information on the description of the substances, the level of contamination of the aquatic environment, the persistence of the substances in the aquatic environment and other media, the toxicity (mainly in the aquatic environment) and bioaccumulation. On the basis of the available data, water quality standards are proposed.

The study is presented in four parts:

Part I	Benzene, ethylbenzene and isopropylbenzene
Part II	Chlorinated benzenes
Part III	Chloronitrobenzenes and 4-chloro-2-nitroaniline
Part IV	Chloroanilines

The study has been carried out by F. Balk, P.C. Okkerman, M. Hof, A. van de Bovekamp and J.W. Dogger. Critical remarks of J. Blok were greatly appreciated.

SUMMARY

The impact of benzene, ethylbenzene and isopropylbenzene on the aquatic environment is evaluated in order to provide a toxicological basis to derive a proposal for water quality criteria. This evaluation is an update of two earlier reports to EC (Persoone and Vanhaecke, 1982 and Chambon, 1984).
Data on physical and chemical properties, degradability, toxicity, chemical detection and environmental concentrations have been reviewed. The availability of data on the different subjects is summarized in Table i.

Benzene, ethylbenzene and isopropylbenzene are widely used for a variety of products in the chemical industry; they may be used as solvents; benzene and isopropylbenzene are components of gasoline.
Total production in the EC in 1987 of benzene, ethylbenzene and isopropylbenzene is estimated at $>440*10^3$, $2900*10^3$ and $<1370*10^3$ tons/year, respectively.

Benzenes can be chemically analyzed by infrared spectroscopy and by gas chromatography. Generally the substances are detected by GC-MS or GC-FID. Detection limits depend on the technique and on the complexity of the sample, but may be less than 1 ng/l.

Recent data on measured concentrations in air, water and soil are lacking. Common concentration of benzene in river water were 0.2 to 3 µg/l. Mean concentrations of ethylbenzene in the river Rhine were 0.3 µg/l with peaks to 2 µg/l. Concentrations in contaminated areas may rise to > 5 mg/kg soil or to > 5 µg/l in groundwater.

As the log K_{ow} is low, accumulation on sediment and in animal tissue is low, . The bioconcentration factor of benzene for *Daphnia pulex* is 200 and for fish <10. The chemical reactivity of benzenes in the aquatic environment is low, due to the high stability of the benzene ring system. Benzene and isopropylbenzene are classified as readily biodegradable and also ethylbenzene is completely biodegradable under aerobic conditions. Biodegradation is an important route of elimination from the aquatic system.
Another important route of elimination is volatilization. Calculated half-lives for volatilization from a "standard water system" were less than 6 hours. In air they will degrade rapidly by indirect photolysis with radicals.

Aquatic toxicity data for benzene shows a large variation, with as extremes acute LC_{50} of 5.3 and 420 mg/l for two fish species. The reproducibility of test results is low: the LC_{50} in two tests with *Daphnia pulex* differed by a factor 20. The variability in test results may be due, at least partly, to the high volatility of benzene. Information on chronic toxicity is lacking. Studies on early life stages of two marine species showed that embryo development was affected already at 0.05 mg/l.
The acute toxicity of ethylbenzene is comparable to the toxicity of benzene. No chronic toxicity studies were found.
Toxicity data for isopropyl are limited to a mollusc and two crustacean species. The same variability in test results is observed and the toxicity seems to be comparable to the toxicity of ethylbenzene. QSAR estimations of the toxicity based on log K_{ow} did not predict the toxicity of ethylbenzene very well, and therefore the QSAR was not considered to be sufficiently reliable to estimate the toxicity of isopropylbenzene.

The $NOEC_{ecosystem}$ was estimated by the modified EPA method: the lowest NOEC for benzene was divided by 10 to arrive at a maximum tolerable concentration of 0.005 mg/l. The toxic effects of benzene, ethylbenzene and isopropylbenzene are expected to be additive. Therefore a water quality criterion of 5 µg/l is proposed for the sum of the three benzenes. This level comes close to the environmental levels cited in the earlier reports as "not uncommon" in river water for benzene and ethylbenzene.

Standards proposed by the USA and Canada for benzene and ethylbenzene range between 2.4 and 5 µg/l and are comparable with the proposed water quality standards (5 µg/l).

Data on mammalian toxicity are incomplete. A tentative "Tolerable Daily Intake" of 170 µg/kg body weight/day was proposed for benzene and 136 µg/kg body weight/day for ethylbenzene. Benzene is considered to have genotoxic properties; it shows carcinogenic activity in man. For ethylbenzene and isopropylbenzene no data are available.

Based on the proposed water quality standard of 5 µg/l a maximum daily intake by man is estimated at 0.33 µg/kg bodyweight/day, which is far below the above mentioned tolerable daily intake levels.

Table i Information available for benzene, ethylbenzene and isopropylbenzene on the different subjects.

Subject:	BENZENE	ETHYL-BENZENE	ISOPROPYL-BENZENE
1. PHYSICO-CHEMICAL CHARACTERISTICS	+	+	+
Production levels	+	+	+
2. ANALYTICAL DETECTION TECHNIQUES			
Detection methods	+	+	+
Detection levels	+	+	+
3. ENVIRONMENTAL LEVELS			
Residues in the atmosphere	±	±	-
Residues in soil and groundwater	±;±	-;-	-;-
Residues in surface water and sediment	±;-	±;±	-;-
Residues in aquatic organisms	-	±	-
Residues in terrestrial organisms	-	-	-
4. PERSISTENCE AND DEGRADATION PATHWAYS			
Abiotic degradation	±	±	±
Biological degradation and metabolism	+	+	+
5. DISTRIBUTION BETWEEN ENVIRONMENTAL COMPARTMENTS			
Volatization	+	+	+
Sorption	+	+	+
Bioaccumulation:			
Aquatic organisms	+	-	-
Terrestrial organisms	-	-	-
Biomagnification	+	±	±
6. TOXICITY			
Aquatic toxicity:			
Acute toxicity (freshwater; marine)	+;±	+;+	±;±
Chronic toxicity (freshwater; marine)	-;-	-;-	-;-
Quantitative structure activity relations (QSARs)	+	+	+
Toxicity to terrestrial organisms	-	-	-
(semi)field studies	-	-	-
Toxicity to mammals (acute; chronic)	+;±	+;±	+;±
Carcinogenic, mutagenic and teratogenic effects	+;±;-	-;-;-	-;-;-

\+ data available
± insufficient data available
\- no data available

ZUSAMMENFASSUNG

Die Wirkung von Benzol, Ethylbenzol und Isopropylbenzol auf die aquatische Umwelt wurde ermittelt mit der Absicht eine toxikologische Grundlage zu beschaffen auf welcher ein Vorschlag für Wasserqualitätskriterien abgeleitet werden könnte.
Dieser Studie stellt eine Aufwertung dar von zwei früheren Berichten an die EG (Persoone und Vanhaecke, 1982 und Chambon, 1984). Werte bezüglich physischen und chemischen Eigenschaften, Abbaubarkeit, Toxizität, chemischer Determinierung und Umweltkonzentrationen wurden untersucht. In Tafel i sind die Ergebnissen dargestellt über die verfügbare Daten der verschiedenen Gegenstände.

Benzol, Ethylbenzol und Isopropylbenzol finden eine breite Anwendung für einen Vielfalt von Produkten der chemischen Industrie; sie können verwendet werden als Lösungsmittel; Benzol, Ethylbenzol und Isopropylbenzol sind Komponenten von Benzin. Die totale Produktion von Benzol, Ethylbenzol und Isopropylbenzol in der EG wird eingeschätzt auf $>440*10^3$, $2900*10^3$ und $1370*10^3$ Tonne/Jahr in 1987.
Benzole können chemisch analysiert werden mittels Infrarot-Spektroskopie und durch Gaschromatografie. Im Allgemeinen werden die Stoffen determiniert durch GC-MS oder GC-FID.
Die Determinierungsgrenzwerte sind abhängig von der Anwendungstechnik und von der Komplexizität des Probemusters, können aber weniger als 1 ng/L betragen.

Rezente Werte von gemessenen Konzentrationen in Luft, Wasser und Boden fehlen.
Übliche Konzentrationen von Benzol in Flusswässern betrugen 0,2 bis 3 µg/L. Mittlere Konzentrationswerte von Ethylbenzol im Rhein betrugen 0,3 µg/L mit Höchstwerten bis 2 µg/L. Die Konzentrationen in verunreinigten Gegenden können ansteigen bis > 5 mg/Kg in Böden oder bis > 5 µg/L im Grundwasser.

Akkumulation an Sedimenten und in tierischen Geweben ist niedrig, da die log K_{ow} niedrig ist.
Der Biokonzentrationsfaktor von Benzol bezogen auf Daphnia pulex ist 200 und für Fische < 10. Die chemische reaktivierung von Benzole in der aquatischen Umwelt ist niedrig wegen der hohen Stabilität des Benzol-Ringsystems.
Benzol und Isopropylbenzol sind klassifiziert als einfach biologisch Abbaubar und auch Ethylbenzol ist vollständig biologisch abbaubar unter aeroben Verhältnissen.
Biologische Abbaubarkeit ist eine wichtige Beseitigungsweise aus dem aquatischen System.
Eine andere wichtige Beseitigungsweise ist Verflüchtigung. Berechnete Halbwerte durch Verflüchtigung aus ein "standarisiertes Wassersystem" wurden in weniger als sechs Stunden erreicht.
An der Luft werden sie schnellstens abgebaut durch indirekte Photolyse mit Radikalen.

Die Streuung von aquatischen Toxizitätswerten für Benzol variiert bedeutend, mit akuten LC_{50} zwischen 5,3 und 420 mg/L für zwei Fischarten als äusserste Grenzwerte. Die Reproduzierbarkeit der Versuchsergebnissen ist gering: die LC_{50} bei zwei Versuchen an Daphnia pulex unterschieden sich mit einem Faktor 20.
Die Variationen der Versuchsresultate könnten möglicherweise zum Teil zurückzuführen sein auf der hohen Verflüchtigungsgrad von Benzol.

Fakten über chronische Toxizität fehlen. Untersuchungen an frühe Lebensstadien von zwei Marinearten zeigten, daß die embryonale Entwicklung schon beeinflusst wurde bei 0,05 mg/L.
Die akute Toxizität von Ethylbenzol ist vergleichbar mit der Toxizität von Benzol. Es wurden keine Untersuchungsergebnisse über chronischer Toxizität gefunden.
Toxikologische Werte über Isopropyl sind beschränkt auf eine Molluscart und zwei Krustazäenarten. Die gleichen Variationen in den Versuchsergebnissen wurden festgestellt und die Toxizität scheint vergleichbar zu sein mit der Toxizität von Ethylbenzol.
QSAR-Einschätzungen der Toxizität begründet auf log K_{ow} ergaben keine deutliche Prognose der Toxizität von Ethylbenzol; deswegen wurde der QSAR nicht als ausreichend zuverlässig betrachtet um die Toxizität von Isopropylbenzol einzuschätzen.

Das $NOEC_{Ecosystem}$ wurde eingeschätzt durch die angepasste EPA-Methode; die niedrigste NOEC für Benzol wurde durch 10 geteilt um auf eine maximal akzeptabele Konzentration von 0,005 mg/L zu gelangen.
Die toxischen Effekten durch Benzol, Ethylbenzol und Isopropylbenzol sind erwartungsgemäß additiv. Deswegen wird ein Wasserqualitätskriterium von 5 µg/L vorgeschlagen als die Summe der drei Benzole. Dieses Niveau befindet sich nahe an den Umweltniveaus welche in früheren Berichten als "nicht ausserordentlich" in Flusswässern für Benzol und Ethylbenzol genannt wurden.
Das Wasserqualitätskriterium in den USA und in Canada für Benzol und Ethylbenzol varieren zwischen 2,4 und 5 µg/L und sind vergleichbar mit dem vorgeslagenen Wasserqualitätskriterium (5 µg/L).
Toxizitätswerte über Säugetiere sind unvollständig. Eine vorläufige "akzeptabele tägliche Einnahme" von 170 µg/Kg Körpergewicht/Tag wurde vorgeschlagen für Benzol und 136 µg/Kg Körpergewicht/Tag für Ethylbenzol.
Benzol wird betrachtet genotoxische Eigenschaften zu enthalten und deutet auf karzinogenische Aktivitäten an Menschen.

Für Ethylbenzol und Isopropylbenzol sind keine Werten verfügbar.

Ausgehend vom vorgeschlagenen Wasserqualitätskriterium von 5 µg/L wurde die maximale täglich durch Menschen aufzunehmende Menge eingeschätzt auf 0,33 µg/Kg Körpergewicht/Tag, welcher Wert weit niedriger ist als die vorher genannten akzeptabelen täglichen Einnahmeniveaus.

RÉSUMÉ

L'effet du benzène, de l'éthylbenzène et de l'isopropylbenzène sur l'environnement aquatique a été évalué afin de fournir une base toxicologique d'une proposition concernant les critères de qualité de l'eau. Cette évaluation est un supplément à deux rapports précédents pour la CEE (Persoone en Van Haecke, 1982 et Chambon, 1984). On a rassemblé de l'information sur leurs propriétés physiques et chimiques, la dégradabilité, la toxicité, la détection chimique et les concentrations dans l'environnement. Le tableau i comprend un aperçu de la disponibilité de données concernant ces sujets.

On utilise largement le benzène, l'éthylbenzène et l'isopropylbenzène dans l'industrie chimique. Ils sont utilisés comme dissolvants et le benzène et l'isopropylbenzène sont des composants d'essence.
La production estimée en 1987 dans la CEE de benzène, d'éthylbenzène et d'isopropylbenzène fut respectivement > 440 * 10^3, 2900 * 10^3 et < 1370 * 10^3 tonnes/an.

Les benzènes peuvent être analysés par spectroscopie infrarouge et par CG. Les méthodes de détection d'usage sont la CG-spectrométrie de masse ou CG-détection à ionisateur de flamme. Les limites de détection dépendent de la technique utilisée et de la composition de l'échantillon, mais elles peuvent être en-dessous de 1 ng/l.

Des données de mesures récentes sur les concentrations dans l'air, dans les eaux de surface et dans les sédiments nous manquent.
Des concentrations habituelles de benzènes dans les eaux fluviales étaient de 0,2 jusqu'à 3 µg/l. Des concentrations moyennes d'éthylbenzènes dans le Rhin étaient de 0,3 µg/l avec pointes jusqu'à 2 µg/l. Dans des régions polluées les concentrations dans la terre peuvent être > 5 mg/kg ou dans les eaux souterraines > 5 µg/l.

Comme log K_{ow} est bas, accumulation en sédiments et en tissu animal est minime. Le facteur de bioconcentration de benzènes pour le Daphnia pulex est 200 et pour le poisson < 10. La réactivité chimique de benzènes dans l'environnement aquatique est petite à cause de la grande stabilité du système du cercle benzénique. Le benzène et l'isopropylbenzène ont été classifiés comme aisément biodégradables tandis que l'éthylbenzène est aussi complètement biodégradable dans de conditions aérobies. La dégradation biologique est un procédé d'élimination d'importance dans le système aquatique.

Un autre procédé d'élimination important est la volatilisation. La durée de réduction de moitié qu'on a calculés pour la volatilisation d'un "système aquatique standard" est moins de 6 heures. Dans l'air les combinaisons se dégradent vite par moyen de fotolyse indirecte aux radicaux.

Les données sur la toxicité aquatique des benzènes montrent une variation large avec une valeur ultime de CL_{50} pour deux espèces de poisson de 5,3 et 420 mg/l respectivement. La réproductibilité des résultats des tests est minime: deux épreuves avec le Daphnia pulex donnaient des valeurs de CL_{50} qui différaient d'une facteur de 20 l'une de l'autre.

Le manque de conformité entre les résultats peut être expliqué, au moins partiellement, par la grande volatilité du benzène.
L'information sur la toxicité chronique fait défaut. Des études de phases de croissance de deux espèces marines ont montré que le développement embryonal fut influencé déjà à un niveau de concentration de 0,05 mg/l.
La toxicité acute d'éthylbenzène est comparable à celle de benzène. Nous n'avons pas trouvé de recherches sur la toxicité chronique.
Des données de la toxicité d'isopropylbenzène sont limitées à celles d'une moule et de deux espèces de crustacés. Elles montrent la même variabilité de résultats des tests et la toxicité semble être comparable à celle d'éthylbenzène.

Des estimations de la toxicité d'éthylbenzène par la Relation Quantitative de la Structure et l'Activité (RQSA), basées sur log K_{ow}, ne prédirent pas très bien la réalité; c'est pourquoi le RQSA ne fut pas considéré comme suffisamment fiable de estimer la toxicité d'isopropylbenzène.

$NOEC_{écosystème}$ a été évalué par la méthode d'EPA modifiée: le NOEC le plus bas pour le benzène fut divisé par dix ce qui eut pour résultat une concentration acceptable au maximum de 0,005 mg/l. On s'attend à ce que les effets de benzène, d'éthylbenzène et d'isopropylbenzène soient additifs. C'est pourquoi on propose une norme de qualité aquatique de 5 µg/l pour l'ensemble des trois benzènes. Cette norme approche celle dont des rapports précédents mentionnent qu'elles ne sont pas singulières pour le benzène et l'éthylbenzène dans les eaux fluviales.

Les normes proposées par les Etats-Unis et le Canada pour le benzène et l'éthylbenzène varient entre 2,4 et 5 µg/l et elles sont comparables à la norme proposée pour la qualité aquatique (5 µg/l).
Les données sur la toxicité pour les mammifères sont incomplètes. Une "prise acceptable par jour" provisoire de 170 µg/kg poids/jour a été proposée pour le benzène et de 136 µg/kg poids/jour pour l'ethylbenzène. On présume que le benzène ait des propriétes génotoxiques; il est cancérogène pour l'homme.
On ne dispose pas de données sur l'ethylbenzène et l'isopropylbenzène.

Si l'on se base sur la norme de qualité aquatique proposée de 5 µg/l la prise maximale par jour pour l'homme sera estimée à 0,33 µg/kg poids/jour ce qui est beaucoup plus bas que les normes citées ci-dessus pour la prise acceptable par jour.

SAMENVATTING

Het effect van benzeen, ethylbenzeen en isopropylbenzeen op het aquatisch milieu is geëvalueerd om een toxicologische basis te verschaffen aan een voorstel voor waterkwaliteitsnormen. Deze evaluatie vormt een aanvulling op twee eerdere rapporten voor de EEG (Persoone en Vanhaecke, 1982 en Chambon, 1984).
Informatie is verzameld aangaande hun fysische en chemische eigenschappen, afbreekbaarheid, toxiciteit, chemische detectie en concentraties in het milieu. Tabel i bevat een overzicht van de beschikbaarheid van gegevens over deze onderwerpen.

Benzeen, ethylbenzeen en isopropylbenzeen vinden uitgebreide toepassing in de chemische industrie. Ze worden gebruikt als oplosmiddel, en benzeen en isopropylbenzeen zijn bestanddelen van benzine.
De geschatte totale produktie in 1987 in de EG van benzeen, ethylbenzeen en isopropylbenzeen was respectievelijk $>440*10^3$, $2900*10^3$, en $<1370*10^3$ ton/jaar.

Benzenen kunnen chemisch geanalyseerd worden met infraroodspectroscopie en gaschromatografie. Gebruikelijke detectiemethoden zijn GC-MS of GC-FID. Detectiegrenzen hangen af van de gevolgde techniek en van de samenstelling van het monster, maar kunnen lager liggen dan 1 ng/l.

Recente meetgegevens over concentraties in lucht, water en bodem ontbreken. Gangbare concentraties van benzeen in rivierwater waren 0,2 tot 3 µg/l. Gemiddelde concentraties van ethylbenzeen in de Rijn waren 0,3 µg/l met pieken tot 2 µg/l. In verontreinigde gebieden kunnen de concentraties in de grond oplopen tot > 5 mg/kg en in grondwater tot > 5 µg/l.

Accumulatie in sediment en in dierlijk weefsel is gering: log K_{ow} is laag. De bioconcentratiefactor van benzeen voor *Daphnia pulex* is 200 en voor vis <10. De chemische reactiviteit van benzenen in het aquatisch milieu is klein vanwege de grote stabiliteit van het benzeenring systeem. Benzeen en isopropylbenzeen zijn geclassificeerd als goed biologisch afbreekbaar, terwijl onder aerobe condities ook ethylbenzeen volledig biologisch afbreekbaar is. Biologische afbraak is een belangrijk eliminatieproces in het aquatisch systeem.
Een ander belangrijk eliminatieproces is vervluchtiging. Berekende halfwaardetijden voor vervluchtiging uit een "standaard watersysteem" liggen lager dan 6 uur. In de lucht breken de verbindingen snel af via indirecte fotolyse met radicalen.

Aquatische toxiciteits gegevens voor benzenen vertonen grote verschillen, met als uiterste waarden een acute LC_{50} voor twee vissoorten van respectievelijk 5,3 en 420 mg/l. De reproduceerbaarheid van de testresultaten is laag: twee proeven met *Daphnia pulex* leverden LC_{50} waarden op die een factor 20 verschilden.
Het gebrek aan overeenkomst tussen testresultaten kan tenminste gedeeltelijk verklaard worden door de grote vluchtigheid van benzeen. Informatie over chronische toxiciteit ontbreekt. Onderzoek aan twee zeewater soorten lieten zien dat de ontwikkeling van het embryo al werd beïnvloed bij concentraties van 0,05 mg/l.
De acute toxiciteit van ethylbenzeen is vergelijkbaar met die van benzeen. Er zijn geen onderzoeken naar chronische toxiciteit gevonden.
Toxiciteitsgegevens voor isopropylbenzeen zijn beperkt tot een mossel en twee kreeftachtigen. Ze laten een zelfde spreiding in testresultaten zien en de toxiciteit lijkt vergelijkbaar met die van ethylbenzeen. QSAR schattingen van de toxiciteit van ethylbenzeen, gebaseerd op de log K_{ow}, gaven geen erg goede voorspelling van

de werkelijkheid; de QSAR werd daarom niet voldoende betrouwbaar geacht voor een toxiciteitsschatting van isopropylbenzeen.
De $NOEC_{ecosysteem}$ is geschat met behulp van de gewijzigde EPA methode: de laagste NOEC voor benzeen werd door tien gedeeld wat resulteerde in een maximum aanvaardbare concentratie van 0,005 mg/l. De effecten van benzeen, ethylbenzeen en isopropylbenzeen zijn naar verwachting additief. Daarom wordt een waterkwaliteitsnorm voorgesteld van 5 µg/l voor de drie benzenen samen. Deze concentratie benadert die waarvan eerdere rapporten meldden dat ze "niet ongebruikelijk" zijn voor benzeen en ethylbenzeen in rivierwater.
Normen die door de VS en Canada zijn voorgesteld voor benzeen en ethylbenzeen variëren tussen 2,4 en 5 µg/l en zijn vergelijkbaar met de voorgestelde waterkwaliteitsnorm (5 µg/l).
Gegevens over de toxiciteit bij zoogdieren zijn onvolledig. Een voorlopige "Aanvaardbare Dagelijkse Inname" van 170 µg/kg lichaamsgewicht/dag was voorgesteld voor benzeen en 136 µg/kg lichaamsgewicht/dag voor ethylbenzeen. Van benzeen wordt aangenomen dat het genotoxische eigenschappen heeft; het is bij de mens kankerverwekkend. Voor ethylbenzeen en isopropylbenzeen zijn geen gegevens beschikbaar.
Uitgaande van de voorgestelde waterkwaliteits norm van 5 µg/l wordt de maximum dagelijkse inname door de mens geschat op 0,33 µg/kg lichaamsgewicht/dag. Dit is ver beneden de hierboven genoemde waarden voor de aanvaardbare dagelijkse inname.

CONTENTS

CONTENTS (cont'd)

1 DESCRIPTION

Benzene is used as an additive to gasoline and as an intermediate in the synthesis of styrene, synthetic rubber, phenol, cyclohexane, detergents, plastics and resins, nylon and pesticides. It is used as a thinner for lacquers and paints and as a degreasing and cleaning agent. Benzene is also used in dyes, coatings, explosives, flavours and perfumes, photographic chemicals and food processing.

Ethylbenzene is used in the manufacture of styrene and acetophenone, as a solvent in the chemical, paint and rubber industries. It is also used as a constituent of asphalt and naphtha.

Isopropylbenzene is used as a high octane gasoline component; it is used as a thinner for paints and lacquers and it is an important intermediate in phenol manufacture (Sittig, 1981).

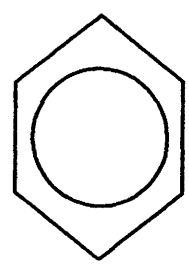
1.1 Identification

benzene

List I Dir. 76/464/EEC	7
EINECS-no.	2007537
CAS no.	71-43-2
Synonyms:	benzeen
	benzol

Molecular formula: C_6H_6

Structural formula:



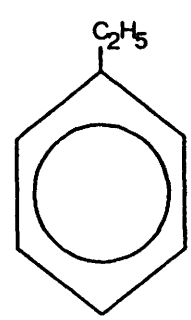
WLN formula: R

ethylbenzene

List I Dir. 76/464/EEC	79
EINECS-no.	2028494
CAS no.	100-41-4
Synonyms:	ethylbenzeen
	ethylbenzol

Molecular formula: $C_6H_5C_2H_5$

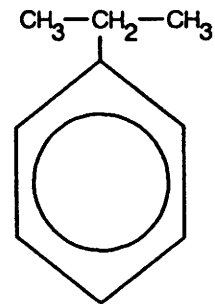
Structural formula:



WLN formula: R2

isopropylbenzene
List I Dir. 76/464/EEC 87
EINECS-no. 2027045
CAS no. 98-82-8
Synonyms: isopropylbenzeen
isopropylbenzol

Molecular formula: $C_6H_5C_3H_7$
Structural formula:



WLN formula: RY1&1

1.2 Physicochemical characteristics

	Benzene	Ethylbenzene	Isopropylbenzene
mol weight	78.12	106.18	120.21
melting point (°C)	5.51	-94.9	-96.0
boiling point (°C)	80.093	136.2	152
density (20°/4°)	0.8794	0.8669	0.864
vapour pressure (mm Hg)	75 (20°C)	10 (25.9°C)	10 mm (38.3°C)
water solubility (20°C) mg/l	1780 [1)]	152	50
volatilization half-life[2)]	5.03	3.1	5.79
" "	4.81		
Log K_{ow}	2.19 [3)]	3.15	3.66

1) Keuning and Janssen, 1987.
2) De Bruin, 1985.
3) De Bruijn, 1991.

1.3 Production levels

Production levels of benzene, ethylbenzene and isopropylbenzene in EC are presented in Table 1.1., derived from EURECO (1990).

Table 1.1 Production levels of benzene, ethylbenzene and isopropylbenzene in EC countries

Country	Capacity in t/y * actual production									Producer
	Benzene			Ethylbenzene			Isoprop.benz.			
Belgium	130000	y90	[1]							Finaneste NV (Antwerp)
	50000	y90	[1]							SA Sopar NV (Zelzate)
	480000	y90	[1]							BASF (Antwerp)
Denmark										
France	330000	y83	[2,3]	260000	(y87)	[2]				CDF Chimie (Carling)
	58000*	y90	[1]							BP Chemical (Laverna)
	150000	(y87)	[2]	280000	(y87)	[2]				Atochem SA (Gonfreville)
	70000	(y87)	[2]							CFR (Gronfreville)
	110000	y83	[3]							Elf Aquitaine (Freyzin)
				6000	(y87)	[2]				Elf (Aquitaine Lacq)
				120000	(y87)	[2]				Progelec (Le-Pont-de-Claix)
				150000	(y87)	[2]				Rhône-Poulenc (Roussillon)
FRG (Germany)	110000	y83	[3]							Deutsche Marathon (Burghausen)
	296000	y90	[1]							VEBA Oel AG (Gelsenkirchen)
	220000	y90	[1]							Redestillationgemeinschaft (Gelsenkirchen)
	200000	y90	[1]							EC Erdoelchemie GmbH (Koeln)
	430000	y90	[1]							Deutsche Shell Chemie GmbH (Godorf)
	195000	y90	[1]							RWE-DEA AG (Wesseling + Heide)
				10000	y90	[1]				RWE-DEA AG (Heide)
				>1000	y90	[1]				BASF AG (Ludwigshafden)
								+	[1]	Hüls AG (Marl)
							340000*	y90	[1]	VEBA Oel (Gelsenkirchen)
Greece										
Ireland										
Italy	750000	y90	[1]							Enmont-Praoil (Prioli+Gela+Porto Torres)
	170000	(y87)	[2]							Saras (Sarroch)
	800	y83	[3]							IPP (Shell) (Rho)
				350000	y90	[1]				Enimont Montedipe SpA (Montava)
				40000	(y87)	[2]				Selm-Societa E. Montedison (Sarroch)
				110000	(y90)	[1]				Enmont-Praoil (Prioli + Sarroch)
							710000	y90	[1]	Enmont-Praoil (Prioli + Porto Terres)
Luxemburg										
Netherlands	815000	(y87)	[2,3]	875000	(y87)	[2]	24000	(y87)	[2]	Dow Chemical BV (Terneuzen)
	285000	y83	[3]							Exxon Chemical Holland (Rotterdam)
	150000	(y87)	[3,4]							DSM NV (Geleen)
				375000	(y87)	[2]				Shell Nederland BV (Moerdijk)
Portugal	50000	y83	[2]							Petrogal (Porto)
					+	[2]				Companihia Nacional de Petroquimica (Sines)
Spain	80000	y90	[1]							Ertoil (Huelva)
	95000	y90	[1]							CEPSA (Algericas)
	80000	y90	[1]							Repsol Petroleo SA (Puertollano)
	15000	y90	[1]							Ensidesa (Avilas)
				120000	y90	[2]				Repsol Quimica SA (Puertollano)
							170000	y90	[1]	Ertisa (Huelva)
United Kingdom	500000	y90	[1]	32000	y90	[1]	150000	y90	[1]	ICI (River Tees)
	450000	y83	[2,3]							Shell (Blessmere Port)
	145000*	y90	[1]							BP (Grangemouth, Wilson)
	170000	y90	[1]							Continental Oil/Conoco (Tamingham)
Total production										
EC	>4400000	(y87)	[2]	2900000	(y87)	[2]	< 1370000	(y87)	[2]	
Total Use EC	6400000	y84	[5]							

1 Information obtained from industry (Eureco, 1990)
2 Water Research Centre (1987)
3 IDET-CEGOS (1983)
4 WHO, 1983
5 Gesellschaft Deutscher Chemiker (1988)

2 ANALYTICAL DETECTION CHARACTERISTICS

2.1 Description of the detection methods

In the previous evaluation on benzene for the EC, Persoone and Vanhaecke (1982) did not report detection methods for benzene.
In another evaluation on ethylbenzene and isopropylbenzene for the EC, Chambon (1984) reported on the detection methods of ethylbenzene and isopropylbenzene and other aromatic hydrocarbons. Analytical methods are:

- gas chromatography (GC)
- headspace method: detection with photo-ionization detector (PID), flame ionization detector (FID) or mass spectrometry (MS)
  detection limit: 0.1 to 1 µg/l
- direct adsorption: onto a solid phase (e.g. XAD-2 or XAD-7); detection with FID or MS
  detection limit: a few ng to 15 mg/l
- solvent extraction: with e.g. freon, hexane, methylene chloride or pentane
  detection limits: 1 ng/l to 50 µg/l
- infrared spectroscopy (I.R.)
- e.g. extraction with freon
  detection limit: 4 to 40 mg/l

Detection in air:

A low level can be detected by filtration of air on charcoal followed by elution with carbon disulphide and detection by GC.

Detection in soil:

First, the soil is mixed with freon; after separation the freon is distilled at 70°C. The residue is dissolved in carbon disulphide. Detection by GC or IR.

Detection in fish:

A mixed fish sample is vacuum distilled. The volatile substances are trapped on a column which is thermally desorbed. Volatile substances are sent onto a GC capillary column.

Detection in water:
The EPA method 602 for purgeable aromates (EPA, 1984) describes a purge-and-trap chromatographic technique for benzene, chlorobenzene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, 1,4-dichlorobenzene, ethylbenzene and toluene in 6 types of water: distilled water, drinking water, surface water, and 3 types of waste water. An inert gas is purged through a contaminated water sample in a chamber with a special temperature. Subsequently the aromates are transferred to the vapour phase and are trapped in a sorbent trap. After purging is completed, the trap is heated and backflushed with the inert gas to desorb the aromates onto a gas chromatographic column. The gas chromatograph is temperature-programmed to separate the aromates which are detected with a photo-ionization detector.
The analytical conditions are:

- temperature-programme: 50°C-2 min., 6°/min to 90°C
- carrier gas: N_2 (20-40 ml/min)
- column packing: 5% SP-1200/1.75% Bentone 34

The detection limits for the EPA method 602 are 0.2 µg/l for both benzene and ethylbenzene with a standard deviations of 10% for benzene and 12% for ethylbenzene.

Geyer (1980) described a detection method for volatile liquid hydrocarbons in seawater, like aldehydes, and aromatic and chlorinated hydrocarbons.
The organics are continuously stripped from a heated, 2-litre sample (70°C) by a pure inert gas (helium). The stripped components are trapped on a Tenax-GC column. Stripping efficiencies were found to be nearly 100%. After stripping, the components trapped on the Tenax-GC, are desorbed by an on-line heating unit (250°C for 15 min.), located outside the GC oven. A pre-column is necessary, since the absorbed compounds are not released simultaneously upon heating. The analysis can be carried out with a GC, provided with the appropriate detector.
The component sensitivity of the entire stripping and analytical method is below 1 ng/l (with a HP 5700A GC with dual FID). The boiling points of the components measured range from about 70-250°C.

2.2 Conclusions

The most frequently applied technique for the detection of benzene, ethylbenzene and isopropylbenzene is preceded by a purge and trap technique. As environmental samples often contain a mixture of substances that can disturb the identification of single substances, this pretreatment is important. Generally, the substances are detected by GC-MS or GC-FID.
Detection limits depend on the applied technique and on the complexity of the mixture. In some cases the detection limit can be less than 1 ng/l.

3 ENVIRONMENTAL LEVELS

In this chapter a review will be given of the concentration levels for benzene, ethylbenzene and isopropylbenzene in various environmental compartments. These levels depend on local environmental conditions like climate, the organic content of sediments, water flow rate of the rivers, etc. Therefore these levels will give only restricted information of the concentration levels that may occur at other locations.

3.1 Residues in various environmental compartments

3.1.1 Residues in the atmosphere

In the previous evaluation for the EC, Persoone and Vanhaecke (1982) reported benzene concentrations of 1 $\mu g/m^3$ to 50 $\mu g/m^3$ in air.

In another evaluation for the EC, Chambon (1984) summarized the concentration levels found for ethylbenzene in air. A summary of these levels is presented in Table 3.1.1.

Table 3.1.1 Concentration levels of ethylbenzene in air (Chambon (1984).

Location	Level ($\mu g/m^{3)}$
US urban air	22
tunnel in Amsterdam	13
car park (USA)	480
motorway (USA)	400
The Hague	240
landfill (UK)	3.6 - 49
"	89

No new data of environmental levels in the atmosphere are available.

3.1.2 Residues in soil

Persoone and Vanhaecke (1982) found that data on benzene levels in sediments are scarce and do not permit any interpretation. High concentration in sediments are not expected to occur because of the relatively low sediment/water partition coefficient.
Background concentrations of benzene in soil in The Netherlands varied between <0.01 and 0.06 µg/kg, but locally concentrations >5 mg/kg soil were observed on contaminated sites (Slooff, 1988). Measured concentrations in groundwater are usually between <0.0005 and 0.03 µg/l, but locally concentrations in groundwater may be >5 µg/l.

In his evaluation Chambon (1984) reported a concentration of 0.5 µg/kg (wet weight) for ethylbenzene in sediment from the Los Angeles county.

No new data have been found for concentration level of benzene, ethylbenzene and isopropylbenzene in soil.

3.1.3 Residues in water

In their evaluation for the EC, Persoone and Vanhaecke (1982) reported concentration levels of benzene in different types of water. These data are presented in Table 3.1.2.

Table 3.1.2 Concentration of benzene in different types of water (Persoone and Vanhaecke, 1982)

Type of water	Concentration (µg/l)
drinking water	
- not uncommon	< 0.1 - 1
- maximum	10
river water	
- not uncommon	0.2 - 3
- maximum	100
seawater	
- not uncommon	< 0.05 - 0.2
- maximum	1 - 4
rain water	0.1 - 0.5

In his evaluation for the EC Chambon (1984) reported on the occurrence of ethylbenzene and isopropylbenzene in water. In 1979 ethylbenzene was detected in the river Rhine in levels between 0.7 and 2.3 µg/l (ECETOC, 1988). Chambon (1984) cites a mean level of 0.3 with peaks to 2 µg/l in the Rhine in 1983. Some of the studies on ethylbenzene report that isopropylbenzene was not detected.

Most references report high concentrations in the neighbourhood of gas works.
Ethylbenzene is detected in seawater in the neighbourhood of oil platforms (up to 5.5 g/kg). Therefore oil platforms can severely pollute the seawater.
On the other hand it was shown that ethylbenzene and isopropylbenzene hardly occur in drinking water.

No new data are available for the levels in water.

3.1.4 Residues in aquatic organisms

In his literature study for the EC, Chambon (1984) reported some concentrations in fish from wastewater. Dover sole and croaker contained 4 µg ethylbenzene/kg in the liver. Also in crab traces of ethylbenzene were detected.

No new data are available in aquatic organisms.

3.1.5 Residues in terrestrial organisms

No data are available on the concentration levels of benzene, ethylbenzene and isopropylbenzene in terrestrial organisms.

3.2 Conclusions

No recent new data have been found on environmental levels of benzene, ethylbenzene and isopropylbenzene in water, sediment and aquatic organisms. The concentrations of ethylbenzene in air are elevated in cities and locations with much traffic. Benzene concentrations in water and soil are usually low but in contaminated areas measured concentrations may rise to >5 mg/kg in soil or to >5 µg/l in groundwater.

In general, the lack of data does not allow a sufficient presentation of concentrations of benzene, ethylbenzene an isopropylbenzene in the environment.

4 PERSISTENCE AND DEGRADATION PATHWAYS

4.1 Abiotic degradation in the aquatic environment

4.1.1 Abiotic degradation

In a previous evaluation for the EC, Persoone and Vanhaecke (1982) reported two references concerning the chemical degradation of benzene in the aquatic environment. They confirmed the minor importance of chemical degradation in the elimination of benzene from the aquatic environment when related to the rapid volatilization of benzene.

In another evaluation for the EC, Chambon (1984) reported that ethylbenzene can be readily oxidized in different solutes by molecular oxygen. Water, however, inhibits this oxidation.

The chemical reactivity of benzene is low, primarily because of the resonance stability of the benzene ring system. As benzene absorbs light at wavelengths below 290 nm, direct photolysis in the watercolumn is not to be expected. Likewise, hydrolysis and oxidation are not expected to be important in natural surface water. In air the benzene derivatives will degrade rapidly by indirect photolysis with radical in the upper layer of the atmosphere (EPA, 1981).

Mill et al. (1980) reported that isopropylbenzene is degraded by alkylperoxyl and hydroxyl free radicals into 2- and 4-isopropylphenol in a 30/70 ratio. In the aquatic environment hydroxyl radicals can be formed from other organic substances, (e.g. riboflavin) and by irradiation with sunlight and therefore degradation along this pathway may occur in water.

No literature was available on hydrolysis of isopropylbenzene in water.

4.1.2 Conclusions

Limited information is available on the chemical degradation of benzene, ethylbenzene and isopropylbenzene in the aquatic environment. This may be connected to the low reactivity of these substances under environmental aquatic conditions. Furthermore these substances will volatilize rapidly in air because of their high volatilization rate. In air the substances will degrade rapidly by indirect photolysis with radicals.

4.2 Biological degradation and metabolization

The following literature study is based on a review report on biodegradability of priority chemicals by Keuning and Janssen (1987). In an additional search relevant international literature from 1986 until the beginning of 1991 was screened for publications on the biodegradation of benzene, ethylbenzene or isopropylbenzene.

Standardized tests are available to determine biodegradation under aerobic conditions (Annex V in Dir. 79/831/EEC). However, most of the studies reported in literature do not comply with these standardised methods. Therefore, an attempt has been made to classify the substances in line with definitions used by OECD and EC on the basis of the available data on test conditions and results. The following definitions are used:
Primary degradation is the alteration of the chemical structure resulting in the loss of a specific property, or the loss of the parent compound. Ultimate biodegradation is achieved when the test compound is totally utilized by micro-organisms,

producing carbondioxide, water, mineral salts and new biomass, or mineralization.
A substance is classified as readily biodegradable if it shows a positive result in some defined stringent test methods which provide limited opportunities for biodegradation and acclimatization. Readily biodegradable substances are assumed to undergo rapid and ultimate biodegradation in the environment. Substances that biodegrade under more favourable test conditions (prolonged exposure, high biomass concentration, low substance concentration) may be classified as inherently biodegradable. This does not necessarily mean that mineralization is complete, nor that rapid and reliable biodegradation in the environment will occur.

4.2.1 Benzene

Biodegradation in standard tests

Benzene was classified as readily biodegradable (OECD TG 301C) by MITI (1985).

Biodegradation in the natural environment

Compounds containing an aromatic ring in their structure are wide-spread in nature. Therefore it is not surprising that several native micro-organisms can degrade aromatic compounds like benzene. It is proposed that benzene can be (completely) biodegraded under favourable aerobic conditions in the natural environment (e.g. sufficient nutrients, low concentrations of benzene (< 0,1%) and other (toxic) compounds (Slooff, 1988).

Delfino and Miles (1985) demonstrated that benzene was degraded in groundwater. Under aerobic conditions benzene (1 mg/l) was degraded completely within a period of 16 days. Under anaerobic conditions no degradation was observed within a period of 96 days.

Biodegradation under experimental conditions
Several researchers studied the microbial degradation of benzene and isolated micro-organism that can use benzene as the sole source of carbon and energy, i.e. *Pseudomonas putida*, *Moraxella sp.*, *Nocardia corallina* (Gibson and Subramanian, 1984).

Oldenhuis et al. (1989) described the effect of addition of *Pseudomonas* strain GJ31 to soil slurries (at 30°C) containing approximately 500 µM benzene.
Without inoculation, the indigenous microflora needed about 30 h for adaptation to degrade chlorobenzene and 100 h for complete removal. Inoculation with cultures of strain GJ31 (5 mg dry weight/l slurry) realized rapid degradation and complete disappearance within 50 h. The results showed that degradation was due to growth of the inoculated cells using benzene as source of carbon and energy. Addition of activated sludge did not stimulate degradation. The degradation rate of aromatic solvents by the added bacteria in soil slurries was similar or higher than that observed in liquid cultures of the same organisms.

Meyer et al. (1984) studied benzene degradation with batch experiments in which sediment of an oil refinery settling pond was used as microbial inoculum. Benzene degraded rapidly when dosed separately, but was not metabolized in the presence of phenol (initial concentrations 100 mg/l). Arvin et al. (1989) found similar results for benzene dosed separately or with other aromatic compounds in a mixed substrate.

Karlson and Frankenberger (1989) assessed the biodegradation of benzene (initial

concentration approximately 450 µg/l) in gasoline contaminated groundwater upon amendment with nutrients (NH_4- / NO_3-N: 100 mg/l) and an enriched mixed cocktail of adapted micro-organisms in batch experiments at 23°C. The benzene content slowly degraded in the unamended sample but dramatically dropped in concentration in the N-amended water and in the inoculated plus N-amended water. In both experiments more than 95% of the benzene in groundwater was removed through microbial action within 73,5 h; a threshold concentration of 22-35 µg/l was observed below which no further degradation took place.

Melcer et al. (1989) measured the variability of influent and effluent concentrations of trace contaminants at a full-scale municipal sewage treatment plant, treating combined domestic and industrial wastewaters. The benzene concentration was reduced from a mean concentration of 0,8 µg/l in the influent by approximately 60% to a mean effluent concentration of 0,3 µg/l.

Aerobic metabolism occurs trough the formation of catechol by means of incorporation of molecular oxygen catalyzed by dioxygenases. Subsequently catechols are further degraded by ortho- or meta-cleavage in the endproducts succimic acid and acetyl-CoA (ortho-cleavage) and pyruvic acid and acetaldehyde (meta-cleavage).

Biodegradation of benzene under anaerobic conditions is also possible. Major et al. (1988) investigated the biodegradation of benzene, toluene and xylene isomers (BTX; initial concentration of each compound: 3 mg/l) in anaerobic batch microcosms containing shallow aquifer material at 10°C. After an incubation period of 62 days with nitrate (10 mg/l) the percentage of remaining BTX was 5-15% for the individual compounds compared to the percentage of remaining BTX in the sterile control of 79-86% for the individual compounds. Denitrification was positively confirmed by acetylene blockage of nitrate reductase resulting in accumulation of nitrous oxide.

Hutchins et al. (1991) studied microcosms under anaerobic conditions at 12°C from uncontaminated aquifer material and aquifer material contaminated with JP-4 jet fuel. The microcosms were spiked with nitrate to yield 30 or 75 mg NO_3^-/l and several aromatic compounds (final concentration 2-7 mg/l for each compound). In experiments with uncontaminated material benzene was not biodegraded; ethylbenzene biodegraded after a lag period of approximately 30 days to 0.5 mg/l at the end of the 56-day test period. An identical test with contaminated aquifer material showed no removal of benzene and ethylbenzene during the 160 day test period. Wilson et al. (1986) studied the biotransformation of benzene in methanogenic aquifer material. After an incubation period of 40 and 120 weeks (incubation at 17°C in the dark) the benzene initial concentration of 613 µg/l had decreased to 27 and 4% of the sterile control, respectively.

Vogel and Grbic-Galic (1986) observed mineralization of benzene in a mixed culture under anaerobic, methanogenic conditions at 35°C (in the dark). Benzene (3 mM) was used as the sole carbon and energy source. Phenol was the first metabolite detected; the incorporated oxygen atoms originated from water molecules. In another study of Grbic-Galic and Vogel (1987) anaerobic transformation of benzene by mixed methanogenic cultures was described once more. The experiments with this inoculum (10%) were carried out at 35°C in the dark. Benzene was semi-continuously supplied as carbon and energy source in the defined mineral medium. The cultures were fed unlabelled or ^{14}C-labelled benzene. Gas production was monitored over a period of 60 days. In 4 days the initial

benzene concentration of 15 mM was reduced by 66% to 5 mM with concomitant production of phenol. Complete disappearance of benzene was observed after 34 days. Carbon dioxide and methane were the final products of fermentation. Phenol was found to be the major aromatic intermediate. Additional compounds detected were cyclohexine and propanoic acid.

Schink (1985) carried out methanogenic enrichment experiments using sewage sludge or sediment samples as inoculum (5 ml of anoxic mud per 50 ml enrichment cultures) with hydrocarbons as sole organic substrate. No significant formation of methane was observed in enrichment cultures containing benzene (1 ml/l medium) during 14 weeks of incubation at 28°C and pH 7.2-7.3.

4.2.2 Ethylbenzene

Biodegradation in standard tests

No standard tests on the biodegradation of ethylbenzene were reported in the literature.

Biodegradation in the natural environment

Almost no data are available in literature on the biodegradation of ethylbenzene in the natural environment. Bouwer (1989) performed aerobic, batch studies at 20°C on samples from a strip-pit pond contaminated with alkylbenzenes and chlorobenzene from tank-truck washing operations. Over a 43-day incubation period 2 $\pm$ 3% ethylbenzene (initial concentration 100 µg/l) was biotransformed in pondwater. Ethylbenzene was transformed without an apparent lag time in pond water amended with nutrients (10 mg acetate/l; 5 mg N/l; 5 mg P/l) as concentrations were below the detection limit after 12 days of incubation. The addition of nutrients and sediment collected from the bottom of the strip-pit pond (concentration 0.61 g/l) to the pond water produced similar rapid biotransformation. In a repeated batch experiment with pondwater (with and without sediment) amended with nutrients, ethylbenzene (100 µg/l) was completely biotransformed within 24 hours.

In an experimental aerobic activated sludge installation with an influent concentration of 60 mg/l, Kincannon et al. (1983) observed a removal percentage of more than 99% for ethylbenzene. Evaporation and adsorption to activated sludge were responsible for a removal of 5.2-7.5% and 0-0.19%, respectively. Apparently the remaining 92.4-94.6% was removed by microbial activity.

Biodegradation under experimental conditions

Biodegradation of ethylbenzene under aerobic conditions by several micro-organisms is described. Gibson (1972) already described the metabolism via 3-ethylcatechol by *Pseudomonas* sp. and a *Nocardia* sp. In other studies by Gibson et al. (1973, 1984) the metabolism of ethylbenzene (100 mg/l) by *Pseudomonas putida* was investigated. The major metabolic route took place via 1,2-dihydroxy-3-ethylcyclohexa-3,5-dieen and cis-1,2-dihydroxy-3-(1-hydroxyethyl)cyclohexa-3,5-dieen followed by ring fission and further metabolism.
The minor route was via the metabolite 1-phenylethanol which was degraded to the corresponding catechol and subsequently transformed via meta ring-fission.

Smith and Ratledge (1989) reported on catabolism of alkylbenzenes by *Pseudomonas* sp. NCIB 10643. *Pseudomonas* sp. NCIB 10643 grew on a range of mono-substituted alkylbenzenes (C_2-C_7), including ethylbenzene and isopropyl-

benzene. Both alkylbenzenes were metabolized via ring attack, proceeding via initial dioxygenase activity resulting in 3-alkyl-substituted catechols subsequently followed by the meta ring-cleavage pathway.

Utkin et al. (1991) isolated *Pseudomonas* sp. Y2 from oil-contaminated soil in the presence of ethylbenzene as a sole carbon and energy source. Strain Y2 transformed ethylbenzene via the metabolites 2-phenylethanol and phenylacetate and also via 1-phenylethanol. Detection of catechol-2,3-dioxygenase in cell-free extracts of strain Y2 indicated that ethylbenzene could be degraded via the corresponding catechols and subsequently the meta ring-cleavage pathway.

Holland et al. (1985) described the metabolism of ethylbenzene (0.5-1 mg/l) by the fungus *Mortierella isabellina*. As metabolite phenylethanol was detected.

Bouwer and McCarty (1985) studied aerobic columns with immobilised micro-organisms growing on 1 mg acetate/l, which degraded low ethylbenzene concentrations (influent concentration 9.1 µg/l). When the columns were operated with a retention time of 20 minutes the removal of ethylbenzene was 99%.

Few data are available on anaerobic degradation of ethylbenzene. Wilson et al. (1986) studied the biotransformation of ethylbenzene in methanogenic aquifer material. After an incubation period of 20 weeks, 20°C in the dark, no degradation of ethylbenzene (269 µg/l) was observed; after 40 weeks of incubation the ethylbenzene concentration had decreased to 69% of the sterile control.

Bouwer and McCarty (1983) observed no transformation of ethylbenzene (40-114 µg/l) under anaerobic denitrifying conditions with activated sludge as inoculum at 25°C during an incubation period of 11 weeks.
Bouwer (1985) described that immobilised micro-organisms in anaerobic methanogenic columns, growing on acetate as the primary substrate could not degrade simultaneously added ethylbenzene (initial concentration 12 µg/l).

4.2.3 Isopropylbenzene

Biodegradation in standard tests

Isopropylbenzene was classified as readily biodegradable (OECD TG 301C) by MITI (1985).

Biodegradation in the natural environment

No data are available in literature on biodegradation of isopropylbenzene in the natural environment. However, as substances containing an aromatic ring structure are wide-spread in nature, it is assumed that isopropylbenzene can be biodegraded under aerobic conditions by a large variety of micro-organisms.

Biodegradation under experimental conditions

Information on the biodegradation of isopropylbenzene under experimental conditions is limited. Based on the simple aromatic structure it is generally assumed that isopropylbenzene is biodegraded under aerobic conditions, but that the isopropyl moiety is hindering microbial oxidation resulting in lower degradation rate. Isopropylbenzene for example was degraded a factor 2 slower than toluene by a toluene-grown culture of *Pseudomonas putida* (Gibson, 1972; Gibson and Subramanian, 1984). Proposed is that degradation occurs via 3-alkyl-substituted catechols.

Smith and Ratledge (1989) reported on catabolism of alkylbenzenes by *Pseudomonas* sp. NCIB 10643. *Pseudomonas* sp. NCIB 10643 grew on a range of mono-substituted alkylbenzenes (C_2-C_7), including ethylbenzene and isopropylbenzene. Both alkylbenzenes were metabolized via ring attack, proceeding via initial dioxygenase activity resulting in 3-alkyl-substituted catechols subsequently followed by the meta ring-cleavage pathway.

No data are available in literature on the biodegradation of isopropylbenzene under anaerobic conditions.

4.2.4 Conclusions

As substances containing an aromatic ring structure are wide-spread in nature, it is assumed that substances with a similar structure like benzene, ethylbenzene and isopropylbenzene, are biodegradable by a variety of micro-organisms.

Benzene is classified as readily biodegradable. Anaerobic biodegradation was reported but at a very low rate and after a long lag phase.

Under aerobic conditions ethylbenzene is completely biodegradable. Biodegradation under anaerobic conditions does not seem to be important.

Isopropylbenzene is classified as readily biodegradable. Biodegradation under anaerobic conditions has not been reported.

5 DISTRIBUTION BETWEEN ENVIRONMENTAL COMPARTMENTS

5.1 Volatilization

In a previous evaluation on benzene for the EC, Persoone and Vanhaecke (1982) found an evaporation half-life of 37 minutes in a water column of 1 meter depth at 25°C. They also reported on the rapid volatilization of benzene from oil spills.

In another evaluation for the EC, Chambon (1984) found that ethylbenzene evaporates from water with a half-life of 7.5 hours for a water depth of 1 meter at 25°C. Under the same conditions a half-life of 5.79 hours was reported for isopropylbenzene.

According to Mackay and Leikonen (1975) an evaporation half-life for benzene was 4.81 h at 25°C and 5.03 h at 10°C.

Hellman (1987) studied the vaporization behaviour of benzene. He developed a model for the quantification of the volatilization rate in the environment. With this model experiment a volatilization rate of $11.6*10^{-3}$ min^{-1} was found.

As benzene volatilizes rapidly most degradation takes place in air where it is degraded quickly: t½ < 1 day (Ware 1980).

Ince and Inel (1989) carried out laboratory experiments to measure the volatilization rate of different substances. For ethylbenzene a volatilization (first-order) rate constant of 0.71 h^{-1} (25°C) was found. Aeration of the solution resulted in a doubling of the volatilization rate.

No literature was available on the volatilization of isopropylbenzene from water.

In recent years several models have been developed to estimate the volatilization behaviour of substances in the natural environment. In this evaluation study the estimation method developed by De Bruin (1985) is used to estimate the volatilization half-life ("standard half-life"). The calculation is based on physical parameters like water solubility, vapour pressure, water speed (1 m/s), air velocity (3 m/s) and water depth (1 m). The calculation method is presented in Appendix 1. The calculated half-lives for the benzene, ethylbenzene and isopropylbenzene are presented in Table 5.1.1.

Table 5.1.1 Calculated volatilization rate (standard half-life) for benzene, ethylbenzene and isopropylbenzene, according to De Bruin (1985).

Substance	Half-life (h)
benzene	5.0
ethylbenzene	3.1
isopropylbenzene	5.8

A summary of published data on volatilization rates and half-lives data is given in Table 5.1.2.

Table 5.1.2 Volatilization rates and half-lives reported for benzene, ethylbenzene and isopropylbenzene.

Substance	Volatilization rate (h^{-1})	Half-life (h)	Ref.
benzene	1.1	0.62	1
	4.8	0.14	2
	0.70	1.0	3
ethylbenzene	0.092	7.5	4
	0.71	0.98	5
isopropylbenzene	0.12	5.79	4

References:
1 Persoone and Vanhaecke (1982)
2 Mackay and Leikonen (1975)
3 Hellmann (1987)
4 Chambon (1984)
5 Ince and Inel (1989)

Table 5.1.2 shows a high variety in the various volatilization half-lives of each substance. This is mainly caused by the difference in experimental conditions. Differences in half-lives of the various compounds will be caused by the specific physical and chemical properties of the substances
For a proper comparison of the volatility of the substances, Table 5.1.1 should be used, because those calculations are based on identical physical and chemical properties and assumptions.
In general, it can be concluded that the substances will volatilize rapidly from water into the air.

5.2 Sorption

For the fate of a substance in the environment, sorption can play an important role. Substances may adsorb on suspended particles and on sediment where they can remain for a long time. After adsorption a substance may return to the aquatic environment by desorption. It is possible to measure a distribution coefficient of a substance between sediment and water. This parameter is called the sediment/water partition coefficient. The value of this coefficient is dependant on the physical and chemical properties of the substance, e.g. solubility and on the organic carbon content of the sediment. Usually, the partition coefficient is calculated for the carbon content of the sediment. This parameter is called the "organic carbon partition coefficient" (K_{oc}).
For many substances the K_{oc} has been measured. For other substance the K_{oc} can be estimated from the octanol/water partition coefficient (K_{ow}) or from the water solubility.

For benzene, ethylbenzene and isopropylbenzene these partition coefficients and the solubility in water are presented in Table 5.2.1 (data have been derived from chapter 1)

Table 5.2.1 Physical properties of benzene, ethylbenzene and isopropylbenzene

Parameter	Benzene	Ethylbenzene	Isopropylbenzene
solubility (mg/l)	1800	152	50
H (atm·m^3/mol)	$5.4 \cdot 10^{-3}$	$5.8 \cdot 10^{-3}$	$1.28 \cdot 10^{-3}$
log K_{oc}	1.87	1.98	
log BCF	1.34		
vol. half-life (h^{-1})	5.0	3.1	5.8

As a result of the low K_{ow}-value and low K_{oc}-value it is not to be expected that adsorption will play a major role in the distribution in the environment.

5.3 Bioaccumulation

5.3.1 Aquatic organisms

If experimental data are absent, the potential of an organic substance to bioaccumulate may be estimated from its water-octanol partition coefficient, expressed as log K_{ow}, see chapter 1.2. If log K_{ow} is above 3, the substance has the potential to accumulate in the biomass. Whether accumulation actually occurs, depends among other things on the capacity of the organisms to metabolise or eliminate the substance. The log K_{ow} for benzene is 2.2, for ethylbenzene 3.2 and for isopropylbenzene 3.7.

The data collected by Persoone and Vanhaecke (1982) indicate that the accumulation level of benzene in fish is restricted. Extreme values were 0.2 and 135, but most values ranged between 1 and 10. Data for other species were not available at the time.
Only few new data were found for benzene, see Table 5.3.1. All reported bioconcentration factors (BCF) were based on wet weight.
Two BCFs were found for algae: 30 for *Chlorella fusca var. vacuolota* and 2217 for *Selenastrum capricornutum*. Exposure of *Daphnia pulex* to two different concentrations of benzene (40 and 80 µg/l) resulted in BCFs of 225 and 203, respectively. For the fish *Leuciscus idus melanotus* exposure to benzene resulted in a BCF less than 10. Apparently, benzene accumulates (or adsorbs) in algae and in *Daphnia* to a higher level than in the tested fish species. The data confirm the earlier statement in Persoone and Vanhaecke (1982) that bioaccumulation of benzene in fish is not important. Probably the fish is capable to eliminate or metabolize benzene.

For ethylbenzene and isopropylbenzene no data on bioaccumulation were available from the study of Chambon (1984), nor could new data be added in the present update. However, on the basis of the similarity of these substances in chemical characteristics, the similarity in biodegradability and in toxicity (see Chapter 6) it is expected that bioaccumulation in fish will not be important.

Table 5.3.1 Bioconcentrationfactors for benzene

Substance/ species	Lifestage age/size	Conc. (µg/l)	Test system	Uptake period	BCF (wet weight)	Reference
Algae						
Chlorella fusca var. vacuolota		50	S	24 h	30	Freitag, 1985
Selenastrum capricornutum		10000	S	24 h	2217	Casserly, 1983
Crustaceans						
Daphnia pulex	1.9-2.1 mm	40	S	24 h	225	Trucco et al.,
		80	S	24 h	203	1983
Fish						
Leuciscus idus melanotus		50	S	24 h	< 10	Freitag, 1980

5.3.2 Terrestrial organisms

No references were found on bioaccumulation of benzene, ethylbenzene and isopropylbenzene in terrestrial species.

5.4 Environmental distribution

The fate of a substance in the aquatic environment is dependant on a number of physical and chemical characteristics such as volatilization, sorption, bioaccumulation, chemical and biological degradation.
Volatilization, sorption and bioaccumulation have been discussed in this chapter, and chemical and biological degradation was discussed in chapter 4.

After release into the environment benzene, ethylbenzene and isopropylbenzene will mainly partition to the atmosphere because of their high volatilization rate. The bioaccumulation of benzene in fish is not important. Few data are available on adsorption and bioaccumulation of ethylbenzene and isopropylbenzene. The chemical degradation rate in water will be low as a result of the stability of these aromates, but the substances are completely biodegradable in water under aerobic conditions.
Sorption, bioaccumulation and biodegradation, however will be of minor importance because of the short life time in the aquatic environment.

6 TOXICITY

A general remark should be made with regard to the interpretation of the data presented in the literature. Benzene, ethylbenzene and isopropylbenzene are highly volatile substances, indicated by half-lives less than 6 hours (Table 5.1.1). As a consequence these substances will also disappear from the test vessels in toxicity studies. According to Persoone and Vanhaecke (1982) the concentration decrease in static tests is around 25% after 1 day and may rise to 80 - >99% after 4 days, depending on the shape and surface to volume ratio of the container, agitation of the system, the use of a cover and the type of the test (static or flow-through).

As the present report is an update of two EC-reports (Persoone and Vanhaecke, 1982; Chambon, 1984), a search was carried out for toxicity data published since 1980. Since then only a limited number of studies have been reported. Also in most of these new studies, test were static and test substance concentrations have only been reported as nominal concentrations, and should therefore be interpreted with caution. The data are presented in tables 6.1.1 to 6.1.3. A selection of data from the previous EC-reports is included in the tables.

6.1 Aquatic toxicity

6.1.1 Benzene

According to Persoone and Vanhaecke (1982) the toxicity of benzene for micro-organisms varied between 50 and 200 mg/l. However, benzene may interfere with the chemotaxis of micro-organisms at relatively low concentrations, e.g. 0.1 mg/l. However, the chemotaxis inhibition is a reversible process.
No new data were found on the toxicity of benzene to micro-organisms.

The data for algae as reported by Persoone and Vanhaecke (1982) vary largely. In their evaluation they state that maximum no effect levels for marine algae species vary between 10 and 50 mg/l. Compared to this range, the new data for *Ankistrodesmus falcatus* and *Selenastrum capricornutum* seem to be very high. The MATC of < 34 mg/l for the red algae *Champia parvula* (14 days renewal test) falls well within the above mentioned range.

The mean LC_{50}-value for freshwater invertebrates calculated by Persoone and Vanhaecke (1982) approximated 260 mg/l, with the rotatorian as the most sensitive species (14.4 mg/l). Marine crustacean species (mean LC_{50} 67 mg/l) were more sensitive to benzene than the mollusc.
New data were reported for 9 freshwater vertebrates, including two insects, three protozoans, three hydrozoans and a snail. These seem to fit well in this range.
However, also in these data a large intra-species variability in the results is observed (e.g. a factor 20 for the toxicity of *Daphnia pulex*).
For one species extremely sensitive life stages are reported: in tests with the marine echinoderm *Paracentrotus lividus* both the sperm and the embryo were affected at 0.078 mg/l. Endpoints were developmental effects (gut and skeletal mal-formations), fertilization success and chromosomal abnormalities (genotoxic effect). This low value compares well with the value cited by Persoone and Vanhaecke (1982) for the lowest concentration (0.053 mg/l) for "slight" effect in the embryos of a marine fish.

For most fish species the LC_{50} fell within the range of 5 to 50 mg/l (Persoone and Vanhaecke, 1982). Almost no change in LC_{50}-values was found between 24 h and 96 h tests. The new data found in this study for *Oryzias latipes* and *Poecilia reticulata*

seem to be rather high as compared to this range. The sensitivity of two amphibian species tested by the same author were of the same order of magnitude. It is not clear whether benzene concentrations were maintained during these studies.

The new data on benzene include data on a number of new species of freshwater invertebrates and amphibians. The toxicity values fall well within the range of data in the previous report. The lack of studies on chronic toxicity as observed in 1982 is confirmed in the present report. Two studies on early life stages (in an echinoderm and a fish species) showed that the embryo development was affected at concentrations around 0.05 mg/l.

6.1.2 Ethylbenzene

In the previous study on ethylbenzene for EC Chambon (1984) reported toxicity data for bacteria, algae, invertebrates and fish.
The data compare well to the data for benzene, and also the same wide variability is observed which may, at least partly, be connected to the high volatility of the test substance.

Only for two species new toxicity data were reported since 1980. The LC_{50}-values for the crustaceans were 2.1 mg/l for *Daphnia magna* being 40 to 50 times lower than the older values, and 15.4 mg/l for *Artemia*. The latter value fits well into the range of earlier data for marine crustaceans.
Chronic studies with ethylbenzene have not been found.

6.1.3 Isopropylbenzene

In the previous study on isopropylbenzene for EC (Chambon, 1984) toxicity data were available for *Daphnia magna* and *Mytilis edulis*. In addition new data were found on *Daphnia magna* and *Artemia* (Table 6.3.1). The newer data for *D. magna* are more than 2 orders of magnitude lower than the older (0.6 versus 95 mg/l). It should be remarked that the latter concentration (95 mg/l) is higher than water solubility of isopropylbenzene in water: 50 mg/l.
Data on chronic toxicity are limited to an unspecified test with the mussel larvae.

The limited number of data available suggest that the acute toxicity is comparable to the toxicity of ethylbenzene. Chronic data are lacking.

Table 6.1.1 Single species toxicity data for benzene

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness mg $CaCO_3$/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Algae												
Ankistrodesmus falcatus		N	S	Pure	CHU 10 medium	8		24	EC_{50} growth	310	Wong, 1984	2-3
Champia parvula (M)		N	R		Sea water + suppl.			14d	MATC growth, reproduction	<34.2	Thursby et al., 1985	
Selenastrum capricornutum		N	S					96	NOEC growth	600	Slooff, 1983b	
Protozoans												
Tetrahymena ellioti			S					24	NOEC survival	391	Rogerson et al., 1983	
Crustaceans												
Artemia	Larvae		S	Purity >97%				24	LC_{50}	127.3	Abernethy et al., 1986#2	
Daphnia cucullata	≤ 24 h	N	S					48	LC_{50}	373	Slooff, 1983b	
Daphnia magna			S	Purity		6-7		48	LC_{50}	31.2	Bobra, 1983	
Daphnia magna	Neonates		S	Purity >97%				48	LC_{50}	127.3	Abernethy et al., 1986#2	
Daphnia magna	≤ 24 h	N	S					48	NOEC	240	Slooff, 1983b	
Daphnia magna	≤ 24 h	N	S	>97%				48	LC_{50}	400	Slooff, 1983b	
Daphnia pulex	1.9-2.1 mm	A	S					96	LC_{50}	15	Trucco et al., 1983	
Daphnia pulex	≤ 24 h	N	S					48	NOEC	196	Slooff, 1983b	
Daphnia pulex	≤ 24 h	N	S					48	LC_{50}	305	Slooff, 1983b	

Table 6.1.1 Single species toxicity data for benzene - continued

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg $CaCO_3$/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Insects												
Aedes aegypti	3 rd instar	N	S					48	LC_{50}	200	Slooff, 1983b	
Culex pipiens	3 rd instar	N	S					48	LC_{50}	71	Slooff, 1983b	
Hydrozoans												
Hydra oligactis		N	S					48	LC_{50}	34	Slooff,1983a	
Worms												
Erpobdella octoculata		N	S					48	LC_{50}	>320	Slooff, 1983a	
Tubificidae (different species)		N	S					48	LC_{50}	>320	Slooff, 1983a	
Molluscs												
Lymnaea stagnalis	3-4 wks	N	S					48	NOEC	120	Slooff, 1983a	
Lymnaea stagnalis	3-4 wks	N	S					48	LC_{50}	230	Slooff, 1983a	
Echinoderms												
Paracentrotus lividus (M)	Embryo	N	S	Purity >99%				48	EC development	0.078	Pagano, 1988	#2
Paracentrotus lividus (M)	Sperm-offspring	N	S	Purity >99%				48	EC genotoxic	0.078	Pagano, 1988	#2

Table 6.1.1 Single species toxicity data for benzene - continued

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg $CaCO_3$/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Fish												
Carassus auratus	Juveniles	N	S					96	LC_{50}	34.4	Persoone and	
Clupea harengus pallasi (M)	Embryos		S		sal. 2.3%			7d	EC tissue growth delay (reversible)	0.053-0.49	Persoone, 1982	
								5d	EC respiration	0.053		
Clupea harengus pallasi (M)	Spawning females		F		sal. 2.3%			6d	EC embryo development	0.7		
Engraulis mordax (M)	Eggs, 1d-larvae		S		sal. 3%			48	EC development	4-15		
Lepomis macrochirus	Juveniles	N	S					96	LC_{50}	22.5	Persoone, 1982	
Morone saxatilis	Juveniles		F		sal. 2.6%			4w	EC growth	5.3	Persoone, 1982	
Oncorhynchus kisutch	Fry	N	S		Freshwater			96	LC_{50}	8.6	Persoone, 1982	
Oryzias latipes	4-5 wks	N	S					48	NOEC	126	Slooff, 1983b	
Oryzias latipes	4-5 wks	N	S					48	LC_{50}	250	Slooff, 1983b	
Pimephales promelas	Juveniles	N	S		Hard water			96	LOLC	8-9	Persoone, 1982	
Pimephales promelas	Juveniles	N	S		Soft water			96	LC_{50}	33.5	Persoone, 1982	
Pimephales promelas	0.58 g	N	F					48	NOEC	54	Slooff, 1983b	
Pimephales promelas	0.58 g	N	F					48	LC_{50}	84	Slooff, 1983b	
Poecilia reticulata	Juveniles	N	S					96	LC_{50}	36.6	Persoone, 1982	
Poecilia reticulata	3-4 wks	N	S					48	NOEC	265	Slooff, 1983b	
Poecilia reticulata	3-4 wks	N	S					48	LC_{50}	420	Slooff, 1983b	
Salmo gairdneri	Juveniles	N	F					96	LC_{50}	5.3	Persoone, 1982	
Salmo gairdneri	5-8 wks	N	S					48	NOEC	40	Slooff, 1983b	
Salmo gairdneri	5-8 wks	N	S					48	LC_{50}	56	Slooff, 1983b	

Table 6.1.1 Single species toxicity data for benzene - continued

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg $CaCO_3$/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Amphibians												
Ambystoma americanum	3-4 wks	N	S					48	NOEC	120	Slooff, 1983b	
Ambystoma americanum	3-4 wks	N	S					48	LC_{50}	370	Slooff, 1983b	
Xenopus laevis	3-4 wks	N	S					48	NOEC	105	Slooff, 1983b	
Xenopus laevis	3-4 wks	N	S					48	LC_{50}	190	Slooff, 1983b	

Table 6.1.2 Single species toxicity data for ethylbenzene

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg CaCO3/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Bacteria												
Pseudomonas putida								16	LOEC cell number	12	Chambon, 1984	
Algae												
Microcystis aerigunosa								8d	LOEC cell number	33	Chambon, 1984	
Scenedesmus quadricauda								8d	LOEC cell number	160	Chambon, 1984	
Selenastrum capricornutum								96	EC_{50}	>438	Chambon, 1984	
Skeletonema costatum								96	EC_{50}	>438	Chambon, 1984	
Molluscs												
Tapes semidecussata (M)							96		LC_{50} 6.8		Chambon, 1984	
Crustaceans												
Artemia (M)	Larvae		S	Purity >97%				24	LC_{50}	15.4	Abernethy et al., 1986	#2
Daphnia magna			S	Purity >97%		6-7		48	LC_{50}	2.1	Bobra, 1983 #2	
Daphnia magna	Neonates		S	Purity >97%				48	LC_{50}	2.12	Abernethy et al., 1986	#2
Daphnia magna								24	LC_{50}	190	Chambon, 1984	
Daphnia magna								24	LC_{50}	77	Chambon, 1984	
Daphnia magna								48	LC_{50}	75	Chambon, 1984	
Crangon franciscorum (M)									EC_{50}	3.7	Chambon, 1984	
Mysidopsis bahia (M)									EC_{50}	87.6	Chambon, 1984	

Table 6.1.2 Single species toxicity data for ethylbenzene - continued

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg CaCO3/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Nitocra spinipes (M)								24	LC_{50}	16	Chambon, 1984	
Palaemonetes (M)	adult							24	LC_{50}	14.5-17.3	Chambon, 1984	
pugio	larvae									10.2		
Fish												
Carassius auratus								96	LC_{50}	94	Chambon, 1984	
Cyprinodon	Juvenile	N	S	>80%	Seawater			96	LC_{50}	280	Neitmuller, 1981	
variegatus (M)					(filtered)				NOEC	88		
Lepomis macrochirus								96	LC_{50}	150	Chambon, 1984	
Morone saxatilis (M)								96	LC_{50}	0.43	Chambon, 1984	
Pimephales promelas					soft water			96	LC_{50}	48.5	Chambon, 1984	
					hard water				LC_{50}	42.3		
					hard water				NOEC	0.44		
Poecilia reticulata								96	LC_{50}	78	Chambon, 1984	
Salmo gairdneri								96	LC_{50}	14	Chambon, 1984	

Table 6.1.3 Single species toxicity data for isopropylbenzene

Species	Lifestage age/size	A/N	Test system	Purity/ solvent	Testwater	pH	Hardness/ mg $CaCO_3$/l	Expo time (h)	Parameter	Results (mg/l)	Reference	Quality
Molluscs												
Mytilus edulis								25d	EC	1	Chambon, 1984	
Crustaceans												
Artemia (M)	Larvae		S	Purity >97%				24	LC_{50}	13.7	Abernethy et al., 1986	#2
Daphnia magna	Neonates		S	Purity >97%				48	LC_{50}	0.6	Abernethy et al., 1986	#2
Daphnia magna			S	Purity >97%		6-7		48	LC_{50}	0.6	Bobra, 1983	#2
Daphnia magna								24	LC_{50}	95	Chambon, 1984	

(M): marine organism

1 Well performed experiment with reliable test results; sufficiently documented
2 Experiment with reliable test results; insufficiently documented
3 Experiment with unreliable test results

Table 6.1.4 QSAR estimates of the toxicity of benzene, ethylbenzene and isopropylbenzene to various test species (QSARs from Verhaar and Hermens, 1991). See text for discussion on the reliability.

Species	estimated parameter	Benzene (mg/l)	Ethyl-benzene (mg/l)	Isopropyl-benzene (mg/l)
Bacteria	NOEC			
Clostridium botulinum	EC_{50}	641.2	142.2	61.5
Bacillus subtilis	NOEC	289.2	95.5	51.0
Pseudomonas putida	NOEC	77.8		
Microcystis aeruginosa	NOEC	16.0		
Photobacterium phosphoreum	NOEC	76.5	23.1	11.8
Algae				
Skeletonema costatum	EC_{50}	237.7	65.8	32.0
Scenedesmus subspicatus	EC_{50}	120.1	24.4	
Selenastrum capricornutum	EC_{50}	29.7	4.4	1.5
Protozoa				
Tetrahymena pyriformis	EC_{50}	219.2	50.8	22.5
Coelenterates				
Hydra oligactis	NOLC	45.6		
Molluscs				
Lymnaea stagnalis	NOLC	42.6		
Arthropods				
Nitocra spinipes	LC_{50}	110.8	26.9	12.2
Daphnia magna	NOEC	8.2	1.1	0.4
Aedes aegypti	LC_{50}	139.8		
Aedes aegypti	NOLC	91.6		
Culex pipiens	NOLC	53.6		
Fish				
Alburnus alburnus	LC_{50}	135.0	35.0	16.4
Brachydanio rerio	NOEC	14.2	1.8	0.6
Pimephales promelas	LC_{50}	41.8	8.7	3.6
Pimephales promelas	NOEC	4.5	0.6	0.2
P.promelas/B.rerio	NOEC	4.3	0.9	0.4
Poecilia reticulata	LC_{50}	62.7	12.5	5.1
Amphibia				
Ambystoma mexicanum	NOLC	59.6		
Rana catesbeiana	LC50	50.0	10.2	4.2
Rana temporaria	NOLC	54.4		
Xenopus laevis	NOLC	67.9		
Xenopus laevis	LC_{50}	15.5		

6.1.4 Quantitative structure activity relations (QSARs)

If sufficient experimental data for a group of structurally related substances are available, quantitative structure activity relations may be established. For "neutral" substances with a narcotic effect, for 25 species QSARs are available where toxicity is a function of the octanol-water partition coefficient (Verhaar and Hermens, 1991). These QSARs may be applied to estimate the toxicity of benzene, ethylbenzene and isopropylbenzene. Table 6.1.4 presents the toxicity data as estimated by the QSAR approach.

For benzene, a comparison of these estimated toxicity data to the experimental data in Tables 6.1.1 to 6.1.3 can be made for 8 species. For 5 species the QSAR

estimate is within a factor 2 of the experimental data and for 3 species the QSAR estimate is 10 to 30 times lower. For ethylbenzene, the QSAR estimate is within a factor 2 for 3 out of 9 test species and for 6 species the QSAR estimate is 6 to 85 times higher than the experimental data. No comparison could be made for isopropylbenzene.

Although the QSAR approach seems to work for benzene, predictions for ethylbenzene are less reliable. If this would be a trend connected to the molecular structure, the approach cannot be applied to supply a reliable estimate for the toxicity of isopropylbenzene.

6.2 Toxicity to mammals

For benzene, ethylbenzene and isopropylbenzene, the relevant routes for exposure of mammals are through oral intake and inhalation. Therefore, the acute toxicity data as presented in Table 6.2.1 are limited mainly to data for oral and inhalation exposure. The acute toxicity of the substances is relatively low.
Information on chronic toxicity is presented in the following sections.

6.2.1 Benzene

Benzene caused slight to moderate skin and eye irritation and slight to moderate necrosis (ATSDR, 1989). There are indications that benzene causes fetus-toxicity in animals at 160 mg/m^3 air, but no teratogenic effects or effects on reproduction are found (Slooff, 1988; ATSDR, 1989). A tentative "Tolerable Daily Intake" of 170 µg/kg bodyweight/day was derived by Vermeire et al. (1991).

6.2.2 Ethylbenzene

The toxicity of ethylbenzene was evaluated by Janssen and Van der Heiden (1987). Information on chronic oral toxicity is scant. In one study with rats carried out in 1956, ≥ 400 mg/kg body weight per day (application 5 days a week, during 6 months) caused effects on liver and spleen. No effects were observed at 136 mg/kg body weight (Wolf et al., 1956 in Janssen and Van der Heiden, 1987).
Using a safety factor of 1000 (a high factor was used based on the restricted number and less reliable data) a Tolerable Daily Intake (TDI) of 136 µg/kg body weight/day was derived by Vermeire et al. (1991).

A number of acute inhalation experiments also caused effects on the liver, the lowest concentration causing these effects was 1350 mg/m^3. No effects were observed at 218 and 430 mg/m^3 (exposure period 6 h a day, 5 days a week). Using a safety factor of 1000, a toxicologically derived acceptable level in air (for continuous exposure) of 77 µg/m^3 was derived (Vermeire et al., 1991).

6.2.3 Isopropylbenzene

Isopropylbenzene is a CNS depressant. In rats exposed to 500 mg/kg daily for 5 months, hyperemia and congestion were noted in the lungs, liver and kidney. Repeated gastric intubation of 154 mg/kg bodyweight did not affect rats, but at 1462 mg/kg, the kidney weight increased (Chambon, 1984).
No new data on the chronic toxicity of isopropylbenzene were found.

Table 6.2.1 Acute toxicity of benzene (Slooff et al., 1986), ethyl benzene and isopropyl benzene (Sax, 1989)

Compound test organism	Route of administration	Expo. time	Criteria	Result (mg/kg)
Benzene				
rat	oral		LD50	5970
rat	oral		LD50	810
rat	oral		LD50	5600
rat	oral		LD50	3000
rat	oral		LD50	3300
rat	oral		LD50	4900
rat	oral		LD50	930
mouse	oral		LD50	4700
rat	inhalation	4 h	LC50	43,700
rat	inhalation	7 h	LC50	31,950
mouse	inhalation	2 h	LC50	60,000
mouse	inhalation	8 h	LC50	15,000
ethylbenzene				
rat	oral		LD50	3,500
mouse	intraperitoneal		LD50	2,272 *
guinea pig, adult	inhalation		LCLo	10,000 *
human	inhalation	8 h	TCLo Eye,Nerv.,Pulm.	100 *
rat	inhalation	7 h	TCLo Repr.	97 *
rat	inhalation	7 h	TCLo Ter.	985 *
rat	inhalation	7 h	TCLo Ter.	96 *
rat	inhalation	4 h	LCLo	4,000 ***
mouse	inhalation	2 h	LCLo	50
rabbit	skin		LD50	17,800
isopropylbenzene				
rat	oral		LD50	1,400
rat	inhalation	4 h	LC50	8,000 * **
mouse	inhalation	2 h	LC50	24,700
rabbit	skin		LD50	12,300

* mg/l
** mg/m^3
*** g/m^3

Repr. = reproduction effects
Ter. = teratogene effects
Eye = eye effects
Nerv. = nervous-system effects
Pulm. = pulmonary system effects

6.3 Carcinogenicity, mutagenicity and teratogenicity

The Dutch National Institute of Public Health and Environmental Protection (RIVM) has evaluated the effects of benzene, and ethylbenzene for deriving toxicological criteria (Vermeire et al., 1991). The most important results are summarized below.

6.3.1 Benzene

Benzene is considered to have genotoxic properties, although there is no clear evidence of a direct reaction with DNA. In cells inheritable chromosomal abberations causing neoplastical transformation were shown. Benzene was found to be carcinogenic in rodents after oral and inhalation exposure.

Chronic exposure of man to benzene in air at concentrations ≥ 325 mg/m^3 are known to induce leukaemia, mostly initiated with non-neoplasmatic effects on the blood forming system.

Chronic exposure of man to concentrations over 100 mg/m^3 air causes abberations of the hematopoietical system. Effects are anaemia, leucopenia and thrombocytopenia, and in severe cases aplastic anaemia.

Because of its carcinogenic activity in man it is concluded that the risk assessment for benzene should be based on a non-threshold approach. By means of linearly extrapolation of reliable data it is calculated that life-time exposure through inhalation of 12 $\mu g/m^3$ or oral administration of ca. 120 μg per day (100% absorption) causes a risk of 1 : 10^4. As the working mechanism for the occurrence of anaemia in man is unknown, it is assumed that similar concentrations of benzene administered via oral and inhalation exposure causes comparable effects. According to the Dutch Public Health organization the linear model leads to a strong overestimation of the real risk. They conclude that mean concentrations below 1.200 $\mu g/m^3$ (ca. 12.000 μg/day intake) do not exceed the risk of 1 : 10^4.

6.3.2 Ethylbenzene and isopropylbenzene

The carcinogenity, mutagenity and teratogenity of ethylbenzene and isopropylbenzene cannot be evaluated due to a lack of data.

7 ENVIRONMENTAL IMPACT ASSESSMENT

7.1 Comparison of exposure and effects

After release into the environment benzene, ethylbenzene and isopropylbenzene will mainly partition to the atmosphere because of their high volatilization rate. Few data are available on sorption and bioaccumulation. The chemical degradation rate in water will be low as a result of the stability of these structures. However, benzene and isopropylbenzene are classified as readily biodegradable. Therefore, biodegradation in the aquatic environment is expected to occur. In the atmosphere the substances will degrade rapidly by indirect photolysis with radicals.

Observed environmental concentrations for benzene, ethylbenzene and isopropylbenzene were reported to vary from 3 µg/l to below detection limits in river water and seawater. Concentrations in the sediment were unknown.
The relatively low log K_{ow}-values (2.1, 3.2 and 3.8) do not indicate strong bioaccumulative properties. Moreover, experimental data for benzene showed that bioconcentration in fish was lower than predicted on the basis of the lipophility (log K_{ow}). Yet low levels of ethylbenzene were reported in fish (4 µg/kg in the liver) and in crab.

The toxicity of these substances varies roughly between 1 and 200 mg/l. For benzene, this is well below the water solubility, but for ethylbenzene (150 mg/l) and isopropylbenzene (50 mg/l), the water solubility is in the same range.
The lowest reported concentration without effect in an acute test is 40 mg/l for trout. However, effects were observed at lower concentrations in other tests; therefore available No Observed Effect Concentrations cannot be taken as a basis for deriving water quality criteria. An effective concentration for the most sensitive species was 0.08 mg/l in an embryo development test with an echinoderm. The margin of safety between these toxicity data and the above mentioned common environmental levels is at least a factor 25.

7.2 Water quality standard

For the estimation of a 'safe' level for the environment, an extrapolation factor can be applied to the available toxicity data which accounts for the different sensitivity of other, non-tested species in the ecosystem. Various approaches have been proposed to extrapolate from single species toxicity data to a concentration where the aquatic ecosystem is not affected (OECD, 1991). This $NOEC_{ecosystem}$ may also be called maximal tolerable concentration or MTC.
The 'modified US-EPA method' can be applied even if only one LC_{50}-value is available. The method applies an assessment factor of 10 to account for each additional level of uncertainty: (1) from one or several chronic values to the $NOEC_{ecosystem}$, (2) from acute LC_{50}-values for at least algae, Daphnia and fish to a chronic NOEC-value, and (3) from only one acute LC_{50}-value to a chronic value. Thus, the assessment factors are 10, 100 and 1000, respectively.
Another approach is to use the variability in the sensitivity among the various test species as a means to calculate a concentration that is expected to be safe for most (e.g. 95%) of the species in the aquatic ecosystem. In other words, a concentration is calculated that is hazardous for only a small number (5%) of species. Two calculation methods are available, which differ in their basic assumptions regarding the shape of the distribution curve for the species sensitivity. One method assumes a log-logistic distribution (Aldenberg and Slob, 1991), whereas the other assumes a log-normal distribution (Wagner and Løkke, 1991).

The results of the two main approaches are presented in Table 7.2.1. The extrapolation method based on the difference in species sensitivity are very sensitive for extreme values. The inclusion of a NOEC of 600 mg/l for an algae results in an unrealistically low MTC for benzene. Therefore the value calculated by the EPA method (lowest NOEC/10; in this case the NOEC is a LOEC/2) is preferred as MTC: 0.005 mg/l.

Table 7.2.1 Maximum tolerable concentrations for benzene

Extrapolation method	MTC (mg/l)	Remarks
Modified EPA	0.004	lowest NOEC / 10
Aldenberg and Slob, 1991	0.0008	95% protection level, (50% confidence,) 5 test species
Wagner and Løkke, 1991	0.0004	

It is assumed that the toxicity of benzene, ethylbenzene and isopropylbenzene is related to their log K_{ow}. Therefore the toxicity of benzene is expected to be 27 times lower than the toxicity of ethylbenzene and 47 times lower than isopropylbenzene. However, the intraspecies variability for benzene is higher than this difference among substances. Therefore it is proposed to calculate a MTC for benzene and assume that the value is valid for the sum of the three substances.

Another safety factor should account for the combined effects of many other xenobiotic chemicals present in the environment. These benzene substitutes and many other 'industrial' chemicals are neutral substances that show narcosis-type toxicity. The effects of these substances are supposed to be additive (Hermens et al., 1984). The order of magnitude for this safety factor is unknown at the present state of the art. One could suggest a safety factor of 10 when not many other environmental contaminants are expected, but when the environmental load is high, a safety factor of 1000 could be more appropriate.

The magnitude of this safety factor is not only a matter of science but political considerations are involved as well. Therefore no safety factor is applied to the MTC to derive a water quality standard for the sum of benzene, ethylbenzene and isopropylbenzene of 5 µg/l (rounded off). It should be realized that the proposed water quality standard does not take into account that other industrial pollutants may be present at the same time.

This proposed level is detectable by various analytical techniques as described in chapter 2.

For benzene the U.S. has proposed a Maximum Concentration Limit for humans of 5 µg/l. Furthermore a health advisory is proposed at 0.1 µg/l concerning the risk of cancer. For ethylbenzene the U.S. has proposed Maximum Concentration Limit of 700 µg/l for lifetime exposure.
Canadian guidelines (1987) for benzene and ethylbenzene are 5 µg/l and 2.4 µg/l (Joint Research Centre, 1989). Standards of the USA and Canada are comparable with the proposed water quality standards (5 µg/l).
However, the standard proposed by the USA for ethylbenzene of 700 µg/l is more than two orders of a magnitude higher.

In the previous report to EC (Persoone and Vanhaecke, 1982) limit concentrations of 200 and 100 µg/l were proposed to protect freshwater biota and the marine environment against benzene, respectively. In view of the recent approaches in hazard assessment, however, these limits can no longer be maintained.

When the proposed water quality standard is compared to the environmental levels commonly found in the riverwater of 0.2 to 3 µg/l for benzene and 0.7 to 2.3 for ethylbenzene (see Chapter 3.1.3) the levels come close to the proposed water quality standard.

7.3 Human exposure

Exposure of man to substances in the aquatic environment may occur through oral intake from (drinking) water and from fish, shellfish or crayfish. Other possible exposure routes are dermal contact and inhalation. The route of dermal contact is relevant if a substance is lipophilic, whereas inhalation is important for highly volatile substances. For the sake of simplicity, intake by inhalation and dermal contact are considered to be negligible in relation to the oral uptake for benzene, ethylbenzene and isopropylbenzene. The human exposure analysis is therefore exclusively based on oral intake.

For the calculations the total concentration of benzene, ethylbenzene and isopropylbenzene in the water is assumed to be at the level of the proposed water quality standard: 5 µg/l. Assuming a daily water consumption of 2 litre, the oral intake of total benzenes from the water will be 10 µg/day.
The concentration in fish is estimated from the bioconcentration factor ($C_{fish}/C_{water} \leq 10$) to be $\leq$100 µg/kg fish. If furthermore a (relatively high) figure for the daily consumption of fish is assumed to be 100 g, the oral intake will be $\leq$ 10 µg/day.
The total oral intake of total benzenes is estimated at 20 µg/day or for a 60 kg person 0.33 µg/kg body weight/day. Comparison with the "tentative acceptable daily intake" for benzene of 6.5 µg/kg body weight/day and for ethylbenzene of 136 µg/kg body weight/day (see chapter 6.3) the margin of safety is rather high. Therefore the risk of human exposure to benzene, ethylbenzene and isopropylbenzene present in aquatic systems in a concentration of 5 µg/l is expected to be negligible.

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APPENDIX 1.

The calculation method for the volatilization standard half-life according to De Bruin (1985).

In the calculation method some assumptions have been made:

- water flow rate = 1 m/s
- air velocity = 3 m/s
- water depth = 1 m
- temperature = 10 °C

The calculation consists of 6 different steps that will be described below:

1 Henry Constant (H):

$H = p/s$

p = vapour pressure (atm.)
s = water solubility (mol/m^3)
H = Henry Constant ($atm \cdot m^3/mol$)

2 dimensionless Henry constant (H'):

$H' = H/RT$

$RT = 0.024$

3 Liquid-Phase Exchange Constant (k(l)):

if $M < 65$:

$k(l) = 20\sqrt{(44/M)}$

if $M > 65$:

$k(l) = 41.93\sqrt{(32/M)}$

M = molecular weight

4 Gas-Phase Exchange Constant (k(g)):

$k(g) = 4550\sqrt{(18/M)}$

5 Overall liquid-phase mass transfer coefficient (K(L)):

$K(L) = \{H' \cdot k(g) \cdot k(l)\} / \{H' \cdot k(g) + k(l)\}$

6 Half-life (t(½)):

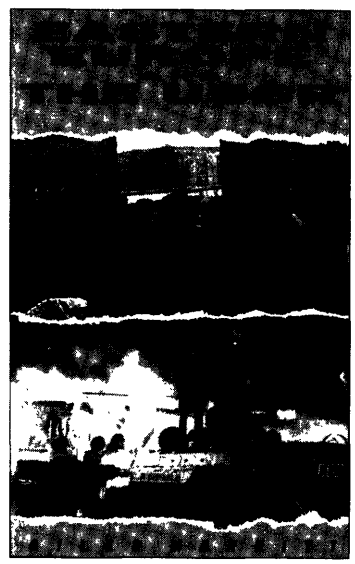
$t(½) = 69.3/K(L)$

t(½) = half-life (hours)

Eastern Europe and the USSR

THE CHALLENGE OF FREEDOM

GILES MERRITT



The sparks of unrest that leapt from Berlin in November 1989 to Moscow's Red Square in August 1991 are firing an explosion of political and economic change. Out of the ashes of Communism is emerging the shape of a vast new European market-place stretching from the Atlantic to the Pacific.

In his fascinating account of Europe's fast-changing East-West relationships, Giles Merritt argues that a massive rescue operation must be mounted to ensure the success of these changes. The upheaval of Communism's collapse is 'The challenge of freedom'.

Written with the cooperation and support of the European Commission, this book sets out to identify the key policy areas where a new partnership is being forged between the countries of Eastern and Western Europe. It offers a privileged insight into the current thinking of European Community officials, politicians and industrial leaders, and analyses the factors that will determine whether the emerging market economies of Eastern Europe can truly be absorbed into a single European economy.

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It is compact, easy to consult (with an index and copious references to the Official Journal and to previous issues), logically structured (to reflect the main fields of Community policy) and wholly reliable. The Bulletin is an essential reference tool, describing the passage of Community legislation through all its stages from presentation of a proposal by the Commission to final enactment by the Council.

Thanks to its topical commentaries on the month's major events, it provides the student of European integration and other interested readers with up-to-date and accurate information about the most recent developments in Community policy — the creation of a single market, economic and social integration, the Community's role in international affairs, etc.

Supplements to the Bulletin are published from time to time, containing important background material on significant issues of the day. Recent Supplements have covered German unification, the Commission's programme for 1992 and European industrial policy for the 1990s.

The Bulletin and its Supplements are produced by the Secretariat-General of the Commission, 200 rue de la Loi, B-1049 Brussels, in the nine official languages of the Community, and can be ordered from the Community sales agents.

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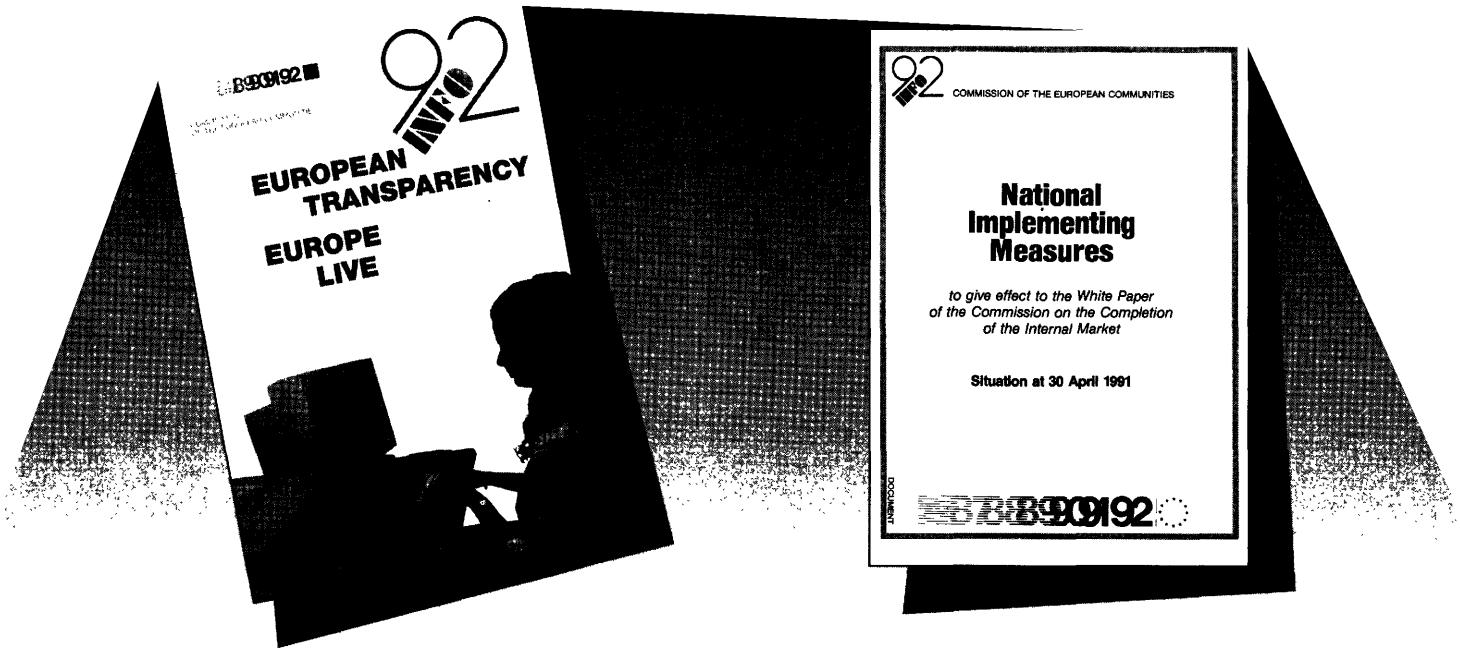

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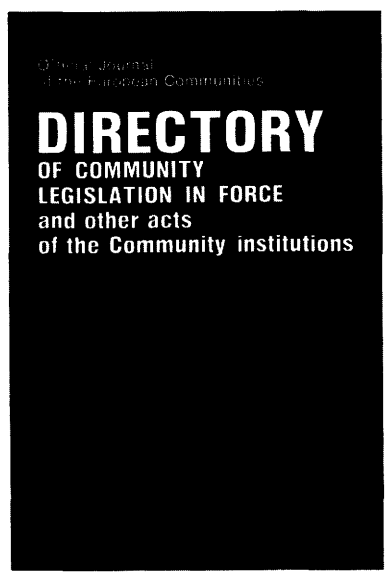

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28/05/93

EUROPEAN ECONOMY

European Economy appears four times a year, in March, May, July and November. It contains important reports and communications from the Commission to the Council and to Parliament on the economic situation and developments, as well as on the borrowing and lending activities of the Community. In addition, *European Economy* presents reports and studies on problems concerning economic policy.

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Unless otherwise indicated, the texts are published under the responsibility of the Directorate-General for Economic and Financial Affairs of the Commission of the European Communities, 200 rue de la Loi, B-1049 Brussels, to which enquiries other than those related to sales and subscriptions should be addressed.

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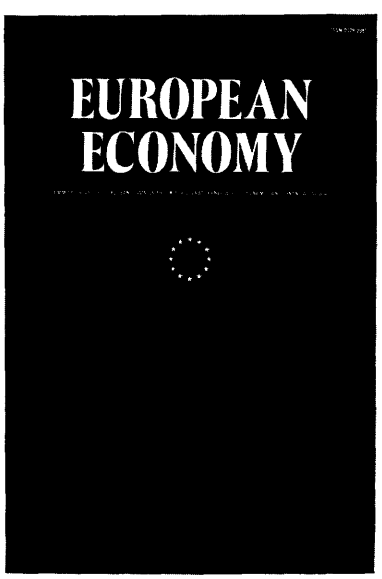

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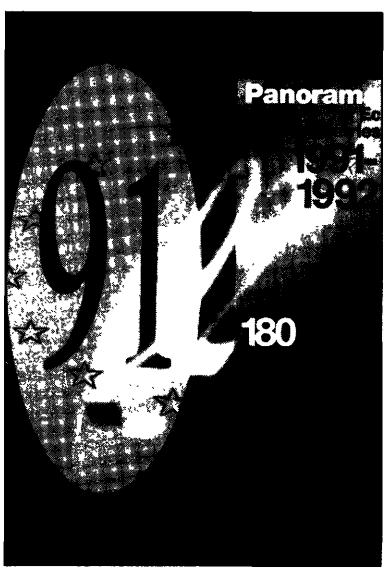

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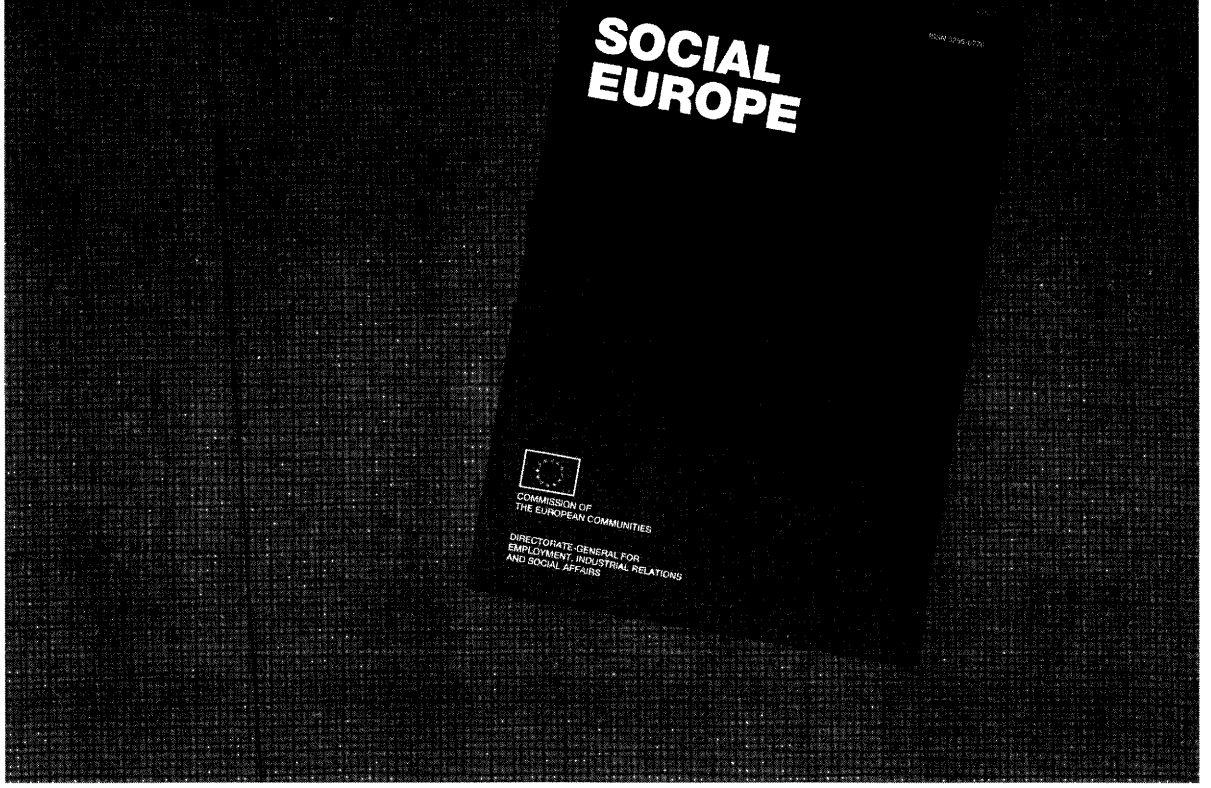

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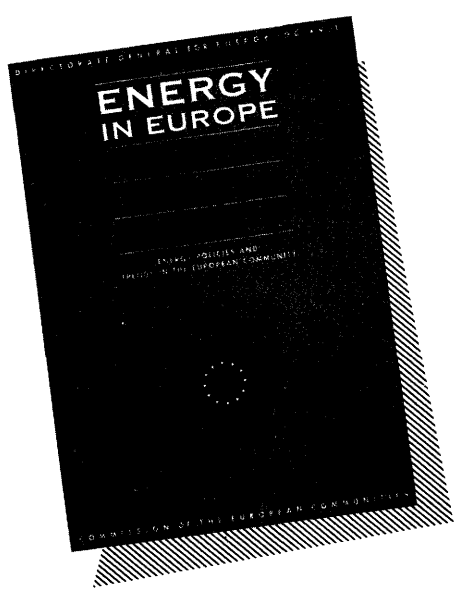

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